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Trioctahedral One-Layer Micas. I. Crystal Structure of a Synthetic Iron Mica*

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The structure of a synthetic iron mica, $\mathrm{KFe_3^{2+}(Fe^{3+}Si_3)O_{10}(OH)_2}$, determined from visually estimated intensities, shows very slight departure from Pabst's idealized biotite structure. The sextuple cell formerly used to describe micas accounts for the observed twinning.

Introduction

The structure determination of synthetic‡ iron mica, 'ferri-annite', $\mathrm{KFe_3^{2+}(Fe^{3+}Si_3)O_{10}(OH)_2}$, was begun (Donnay & Kingman, 1958) at a time when no refined structural data were available for any member of the mica group. To understand the polymorphism of mica, it seemed essential to collect, as accurately as possible, bond lengths and angles for samples of known compositions. For that reason we began our study with an unusual synthetic mica, of relatively simple formula, prepared by Wones (1958) at the Geophysical Laboratory.

In the meantime crystal structures of four natural micas have been published. Xanthophyllite, $\mathrm{Ca_{1\cdot10}Mg_{2\cdot18}Al_{0\cdot72}(Al_{2\cdot95}Si_{1\cdot05})O_{10}(OH)_2}$, a trioctahedral, brittle mica of type 1*M* (Smith & Yoder, 1956), was analyzed by Takéuchi & Sadanaga (1959). Radoslovich (1960) refined the dioctahedral $2M_1$ muscovite, $\mathrm{KAl_2(AlSi_3)O_{10}(OH)_2}$. Zvyagin & Mishchenko (1962), by electron diffraction, studied a trioctahedral 1*M* mica, $\mathrm{KMg_3(AlSi_3)O_{10}(OH)_2}$. An iron-rich phlogopite, $\mathrm{(K_{0\cdot9}Mn_{0\cdot1})Mg_3[(Fe^{3+},Mn)Si_3]O_{10}(OH)_2}$, of type 1*M* was refined by Steinfink (1962). Apart from a slight substitution of Mn for K and $\mathrm{Fe^{3+}}$, this last mica differs from our ferri-annite only in the octahedral layer, which is occupied by Mg instead of $\mathrm{Fe^{2+}}$.

The reader will be able to compare ferri-annite with these four recent structures in the light of recent theoretical discussions by Radoslovich (1959, 1961, 1962, 1963*a–c*) and Radoslovich & Norrish (1962). The crystallochemical implications of our results will be considered in a forthcoming paper.

Experimental

The crystals were prepared by Wones (1958, 1963), who reports on his synthesis as follows: *The crystals formed on the gas-liquid interface of a run containing gas, liquid, wüstite, and fayalite during a slow quench from* 850 °C *at* 2070 *bars total pressure. The optical properties and powder data of these relatively large crystals are identical with those of smaller crystals formed in runs of ferri-annite composition that gave* 100% *yield. The weight proportions of the constituents in these runs are known with* 2% *accuracy, and the composition of the crystals used in this study is estimated to be known with the same accuracy.* The crystals are black, euhedral, platy parallel to (001) and up to 1 mm in thickness. Their (001) cleavage is perfect. Twinning is frequent.

Cell dimensions, determined by single-crystal methods,* are: $a = 5{\cdot}43$, $b = 9{\cdot}40$, $c = 10{\cdot}32$ Å, all three

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‡ Because this composition has not been found in nature, it would be better not to designate it by a mineral name. This is the reason for the quotation marks that enclose ferri-annite, the first time the name appears in the text.

* The cell dimensions have since been refined by Wones (1963) from powder data by least squares: $a = 5{\cdot}430 \pm 0{\cdot}002$, $b = 9{\cdot}404 \pm 0{\cdot}005$, $c = 10{\cdot}341 \pm 0{\cdot}003$, $\beta = 100^\circ\,4' \pm 10'$. The cell volume is 519·9 Å^3 and the calculated density is 3·45 g.cm^{-3}.

$\pm 0{\cdot}2\%$; $\beta = 100^\circ\,0' \pm 10'$. The cell volume is 518·8 Å^3; with $Z=2$ the calculated density is 3·46 g.cm^{-3}. The diffraction aspect is C^*, which is compatible with space groups $C2$, Cm, and $C2/m$.

Untwinned crystals with dimensions about $0{\cdot}2 \times 0{\cdot}2 \times 0{\cdot}03$ mm^3 were selected for precession and Weissenberg work. Fe$K\alpha$ (1·9373 Å) and Mo$K\alpha$ (0·7107 Å) radiations were used, the latter for multiple-film non-integrating Weissenberg exposures about the a axis (zero to 6th layer) and the b axis (zero to 2nd layer). Reflections died out well inside the Mo sphere, so that we arbitrarily selected a limiting sphere of radius 1/1·5 Å^{-1}. It contains 711 independent reflections, 433 of which are actually observed.

Relative intensities were estimated visually with a comparison strip. Weak reflections that could not be estimated were given zero values. Intensities were multiplied by the Lorentz and polarization factors and were corrected for absorption. The relative F_o values thus obtained were later changed to an absolute scale by comparison with the corresponding F_c values. Rogers & Wilson's statistical test (1953), based on three-dimensional data, suggested the presence of a center of symmetry, which is compatible with a negative piezoelectric test performed on the Iitaka (1953) apparatus.

Determination of the crystal structure

In view of the indications we have of a center of symmetry,† we first tried space group $C2/m$. In this space group the general position (j) is 8-fold. It must be chosen for the six Si and two Fe atoms because this is the only way to place these metals at the centers of the tetrahedra. The use of two 4-fold positions is thus ruled out. (Note that we make use of the known $1M$ mica structure.) It follows that Si and Fe^{3+} *must* be in complete disorder in the tetrahedral layers: each site of position (j) accommodates a statistical ion $\mathrm{Si_{0\cdot75}Fe^{3+}_{0\cdot25}}$.

An electron-density projection onto the bc plane was first computed, starting with the coordinates of the biotite structure (Pabst, 1955) and using the atomic scattering factors of *International Tables for X-ray Crystallography* (vol. 3, p. 202). All ions were taken as completely ionized; the scattering factor of OH$^-$ was taken to be equal to that of O^{2-}.

Next all three-dimensional data were used in a least-squares refinement that used only the main diagonal of the matrix. This computation was carried out on the Parametron computer PC–1 at the University of Tokyo. The Busing–Levy program (ORXLS) was then employed for us by Drs Helen Ondik and S. Block at the National Bureau of Standards. Absorption corrections were applied, with the use of a program written by Dr Charles W. Burnham, and Trueblood's full-matrix least-squares program (UCLALS–1) was run on the IBM 7090 computer by Dr D. Koenig at The Johns Hopkins University. A three-dimensional electron-density series was then

Table 2. *Bond lengths and angles, with standard deviations, in the tetrahedral sheet*

The subscripts a, b, *etc.* indicate the sequence in which the equivalent points are listed in *International Tables for X-ray Crystallography* (1952). The $\mathrm{O_I}$ marked c' occurs in the adjacent cell

		Standard deviation
(Si, Fe)–$\mathrm{O_I}$	1·697 Å	0·007 Å
(Si, Fe)–$\mathrm{O}_{\mathrm{II}a}$	1·680	0·013
(Si, Fe)–$\mathrm{O}_{\mathrm{II}f}$	1·670	0·014
(Si, Fe)–$\mathrm{O_{III}}$	1·691	0·011
Average	1·685 Å	
$\mathrm{O_I}$–$\mathrm{O}_{\mathrm{II}a}$	2·770 Å	0·017 Å
$\mathrm{O_I}$–$\mathrm{O}_{\mathrm{II}f}$	2·696	0·016
$\mathrm{O}_{\mathrm{II}a}$–$\mathrm{O}_{\mathrm{II}f}$	2·727	0·003
$\mathrm{O_I}$–$\mathrm{O_{III}}$*	2·778	0·016
$\mathrm{O}_{\mathrm{II}a}$–$\mathrm{O_{III}}$	2·758	0·016
$\mathrm{O}_{\mathrm{II}f}$–$\mathrm{O_{III}}$	2·773	0·016
Average	2·750 Å	
$\mathrm{O_I}$–(Si, Fe)–$\mathrm{O}_{\mathrm{II}a}$	110·21°	0·75°
$\mathrm{O_I}$–(Si, Fe)–$\mathrm{O}_{\mathrm{II}f}$	106·39	0·75
$\mathrm{O}_{\mathrm{II}a}$–(Si, Fe)–$\mathrm{O}_{\mathrm{II}f}$	109·00	0·45
$\mathrm{O_I}$–(Si, Fe)–$\mathrm{O_{III}}$*	110·17	0·63
$\mathrm{O}_{\mathrm{II}a}$–(Si, Fe)–$\mathrm{O_{III}}$	109·84	0·57
$\mathrm{O}_{\mathrm{II}f}$–(Si, Fe)–$\mathrm{O_{III}}$	111·18	0·58
Average	109·47°	
$\mathrm{O}_{\mathrm{II}e}$–$\mathrm{O}_{\mathrm{I}c}$–$\mathrm{O}_{\mathrm{II}f}$	133·62°	0·81°
$\mathrm{O}_{\mathrm{I}c}$–$\mathrm{O}_{\mathrm{II}f}$–$\mathrm{O}_{\mathrm{II}a}$	107·79	0·88
$\mathrm{O}_{\mathrm{II}f}$–$\mathrm{O}_{\mathrm{II}a}$–$\mathrm{O}_{\mathrm{I}c}'$	132·05	0·91
$\mathrm{O}_{\mathrm{II}a}$–$\mathrm{O}_{\mathrm{I}c}'$–$\mathrm{O}_{\mathrm{II}b}$	106·69	0·76

* $\mathrm{O_{III}}$ is the apical O.

Table 1. *Atomic coordinates in synthetic iron mica*

Position	Atom	x/a	$\sigma(x)$	y/b	$\sigma(y)$	z/c	$\sigma(z)$	B
2(b)	K	0	—	$\frac{1}{2}$	—	0	—	2·20
2(c)	$\mathrm{Fe_I^{2+}}$	0	—	0	—	$\frac{1}{2}$	—	0·80
4(h)	$\mathrm{Fe_{II}^{2+}}$	0	—	0·333	0·0004	$\frac{1}{2}$	—	0·88
4(i)	OH	0·634	0·002	0	—	0·401	0·001	0·95
4(i)	$\mathrm{O_I}$	0·015	0·003	0	—	0·166	0·001	1·93
8(j)	$\mathrm{O_{II}}$	0·820	0·002	0·264	0·002	0·167	0·001	2·61
8(j)	$\mathrm{O_{III}}$	0·633	0·002	0·334	0·001	0·391	0·001	1·22
8(j)	(Si, Fe^{3+})	0·575	0·001	0·333	0·0004	0·224	0·0003	0·87

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† In determining the structure of the iron-rich phlogopite, Steinfink (1962) first assumed an acentric space group, then was led to $C2/m$ in the course of the refinement.

synthesized, a program written by Holden & Block for the IBM 7090 being used. All peaks were of expected heights and the background was uniformly low. At this point ferric iron and silicon were considered half-ionized and the final computations were run on the IBM 7094 at The Johns Hopkins University; atomic coordinates were refined by the Busing–Levy program, with individual isotropic temperature factors (Table 1); bond lengths, angles, and their standard deviations (Tables 2, 3, 4) were computed by means of the 'Crystallographic Function and Error' program of Busing *et al.* (ORFFE).

The final R index is 9·3% for the 433 observed reflections. It is 20·8% for all 711 reflections, that is when non-observed reflections (and the weak reflections that were labelled 'non-observed') are included.

Table 3. *Bond lengths and angles, with standard deviations, in the octahedral sheet*

		Standard deviation
Around $\mathrm{Fe_{I}}$, position c, symmetry $2/m$		
$\mathrm{Fe_{I}}-\mathrm{O}_{\mathrm{III}a}$	2·123 Å	0·011 Å
$\mathrm{Fe_{I}}-\mathrm{OH}_{d}$	2·074	0·013
Average	2·107 Å	
$\mathrm{O}_{\mathrm{III}a}-\mathrm{O}_{\mathrm{III}b}$	3·118 Å	0·023 Å
$\mathrm{O}_{\mathrm{III}a}-\mathrm{OH}_{c}$	3·146	0·015
Average	3·137 Å	
$\mathrm{O}_{\mathrm{III}b}-\mathrm{O}_{\mathrm{III}d}$*	2·884 Å	0·021 Å
$\mathrm{O}_{\mathrm{III}a}-\mathrm{OH}_{d}$*	2·779	0·016
Average	2·814 Å	
$\mathrm{O}_{\mathrm{III}a}-\mathrm{Fe_{I}}-\mathrm{OH}_{d}$ $180° - (\mathrm{O}_{\mathrm{III}a}-\mathrm{Fe_{I}}-\mathrm{OH}_{c})$	82·92°	0·37°
$\mathrm{O}_{\mathrm{III}b}-\mathrm{Fe_{I}}-\mathrm{O}_{\mathrm{III}d}$ $180° - (\mathrm{O}_{\mathrm{III}a}-\mathrm{Fe_{I}}-\mathrm{O}_{\mathrm{III}b})$	85·54	0·58
Around $\mathrm{Fe_{II}}$, position h, symmetry 2		
$\mathrm{Fe_{II}}-\mathrm{O}_{\mathrm{III}a}$	2·132 Å	0·011 Å
$\mathrm{Fe_{II}}-\mathrm{O}_{\mathrm{III}f}$	2·112	0·011
$\mathrm{Fe_{II}}-\mathrm{OH}_{a}$	2·075	0·009
Average	2·106 Å	
$\mathrm{O}_{\mathrm{III}a}-\mathrm{O}_{\mathrm{III}f}$	3·143 Å	0·012 Å
$\mathrm{O}_{\mathrm{III}a}-\mathrm{OH}_{a}$	3·143	0·011
$\mathrm{O}_{\mathrm{III}f}-\mathrm{OH}_{a}$	3·119	0·015
Average	3·135 Å	
$\mathrm{O}_{\mathrm{III}a}-\mathrm{O}_{\mathrm{III}c}$*	2·884 Å	0·021 Å
$\mathrm{O}_{\mathrm{III}a}-\mathrm{O}_{\mathrm{III}h}$*	2·866	0·021
$\mathrm{O}_{\mathrm{III}h}-\mathrm{OH}_{a}$*	2·779	0·016
$\mathrm{OH}_{a}-\mathrm{OH}_{b}$*	2·710	0·026
Average	2·814 Å	
$\mathrm{O}_{\mathrm{III}a}-\mathrm{Fe_{II}}-\mathrm{O}_{\mathrm{III}h}$ $180° - (\mathrm{O}_{\mathrm{III}a}-\mathrm{Fe_{II}}-\mathrm{O}_{\mathrm{III}f})$	84·70°	0·41°
$\mathrm{O}_{\mathrm{III}h}-\mathrm{Fe_{II}}-\mathrm{OH}_{a}$ $180° - (\mathrm{O}_{\mathrm{III}f}-\mathrm{Fe_{II}}-\mathrm{OH}_{a})$	83·45	0·46
$\mathrm{OH}_{a}-\mathrm{Fe_{II}}-\mathrm{OH}_{b}$	81·54	0·55
$\mathrm{O}_{\mathrm{III}a}-\mathrm{Fe_{II}}-\mathrm{O}_{\mathrm{III}c}$	85·14	0·60
$\mathrm{O}_{\mathrm{III}a}-\mathrm{Fe_{II}}-\mathrm{OH}_{a}$	96·69	0·42

* Shared edge.

Because the observed intensities are known to only about 15%, further refinement with anisotropic temperature factors is not warranted.

Table 4. *Bond lengths and angles, with standard deviations, in the interlayer*

$\mathrm{K}-\mathrm{O}_{\mathrm{I}c}$	3·015 Å	0·016 Å
$\mathrm{K}-\mathrm{O}_{\mathrm{II}a}$	3·074	0·014
Average	3·054 Å	
$\mathrm{K}-\mathrm{O}_{\mathrm{I}c}'$	3·378 Å	0·015 Å
$\mathrm{K}-\mathrm{O}_{\mathrm{II}f}$	3·332	0·014
Average	3·347 Å	
$\mathrm{O}_{\mathrm{I}c}-\mathrm{O}_{\mathrm{I}d}$	3·393 Å	0·029 Å
$\mathrm{O}_{\mathrm{II}c}-\mathrm{O}_{\mathrm{II}f}$	3·411	0·024
Average	3·402 Å	

The standard deviations are listed as computed even though their significance may be dubious: in his invited paper on *Structure refinement*, at the Sixth International Congress of Crystallography (Rome, 1963) Dr R. E. Marsh pointed out the unreliability of standard deviations in cases where R exceeds 5%.

The orthogonal sextuple cell

The cell used in a preliminary report on ferri-annite (Donnay & Kingman, 1958) is a multiple cell, which can be obtained from our structural cell by the matrix transformation $\bar{1}00/0\bar{1}0/103$. (The inverse matrix, structural cell from multiple cell, is $\bar{1}00/0\bar{1}0/\frac{1}{3}0\frac{1}{3}$.) Not only is it C-centered; it also contains additional nodes at $\frac{1}{3}0\frac{1}{3}$, $\frac{2}{3}0\frac{2}{3}$, $\frac{5}{6}\frac{1}{2}\frac{1}{3}$, $\frac{1}{6}\frac{1}{2}\frac{2}{3}$. Whereas the structural cell is a double cell, the large cell is sextuple. It is very close to being trirectangular.

In reciprocal space the three rows that are quasi-perpendicular are: $[30\bar{1}]^*$, $[010]^*$, and $[001]^*$. The net $(103)^*_0$, which contains $3h,k,\bar{h}$ reflections, coincides nearly (within 6′) with the cleavage plane (001) and is therefore easily photographed on the precession camera. It contains the first two of the above rows and has pronounced pseudohexagonal symmetry. Reflections 060 and $33\bar{1}$ and the symmetry-related ones, $3\bar{3}\bar{1}$, $0\bar{6}0$, $\bar{3}\bar{3}1$, $\bar{3}31$, have equal intensities and outline a hexagon. Only on exceptionally long exposures do other reflections appear without accompanying reflections to simulate hexagonal symmetry. These reflections are 020 and 040. They show that c^* is not a 3- or 6-fold axis of symmetry and that $ll2$ is the symmetry of the net. But if the specimen is twinned by a rotation of 120° about the c^* axis (see discussion below), it could easily be mistaken for a $3T$ mica.

Let us consider the sextuple cell for which the following dimensions were measured from precession films: $a' = 5{\cdot}43$, $b' = 9{\cdot}40$, $c' = 30{\cdot}49$, $\beta' = 90°\,0'$ $(90°\,6'$, if calculated from the β of $100°\,0'$ of the structural cell). Let $h'k'l'$ be the indices of a reflection referred to this large cell. In order to escape destructive interference, the $h'k'l'$ reflections must fulfil the condition $h' + 3k' + 4l' = 0$ mod. 6, which includes the following two: $h' + k' = 0$ mod. 2 and $h' + l' = 0$ mod. 3. In that system of indexing, $h'00$ would appear only as 600, 12,0,0, *etc.* If now we take into account the pseudo-hexagonal character of the *lattice* (not the crystal structure) and apply matrix $100/\frac{\bar{1}}{2}\frac{1}{2}0/\frac{\bar{1}}{2}\frac{\bar{1}}{2}0/001$ to go over to a hexagonal system of coordinates, with indices $HKIL$, the criterion becomes $H + L = 0$ mod. 3. Such criteria should be useful as danger signals. A b-axis zero-layer Weissenberg photograph, on which 200 will appear as a strong reflection, will remove all ambiguity.

Although the sextuple monoclinic cell is, structurally, three layers high, the mica need not be of type $3M$. Ferri-annite, for instance, is a mica of type $1M$, not $3M$ as was erroneously surmised in the preliminary report.

The sextuple cell has been used to describe *all* micas until the advent of X-ray crystallography (see Dana's 'System' of 1892, for example). It offers a special advantage in the description of twinning, as it is the cell of the twin lattice, that is, the multiple lattice (superlattice) whose pseudo-symmetry elements are possible twin elements.

Twinning

Peacock (1943), after Friedel (1904, p. 222), has shown that the mica twin law must be defined by a 180° rotation around a twin axis, [310] or [110], and not by a reflection in a twin plane (110) or (130) (referred to the sextuple cell), because the composition surface is parallel to the cleavage plane (001), which contains the twin axes. Planes (110) and (130) in the sextuple cell become $(33\bar{1})$ and $(39\bar{1})$ in the $1M$ cell.

As to twins by $n.60°$ $(n = 1, 2, \ldots, 5)$ around the axis of hexagonal pseudo-symmetry, [001] in the old cell, [103] in the $1M$ cell, they cannot be distinguished from twins that would result from such rotations around the normal to the cleavage plane (c^* axis in the $1M$ setting) because the angle $[103]:[001]^*$ is 6′ or less. Consider the precession film $(103)^*_0$ again. When the row of the $0k0$ reflections is repeated by a 60° rotation, the same result could have been obtained by a 240° rotation. Likewise 120° and 300° rotations are undistinguishable. It will be shown in the next paragraph that, in the first case, the twin axis can be either $[1\bar{1}0]$ or $[310]$; and in the second case, it can be either $[3\bar{1}0]$ or $[110]$. Finally the appearance of a precession photograph $(103)^*_0$ of a single crystal would not change were the row $[010]^*$ to be repeated by a 180° rotation; this means that the existence of the twin axis [100] cannot be detected on a $(103)^*_0$ photograph.

Let us refer ferri-annite to the $1M$ cell. Five possible twin operations can be described as rotations around $c^*[001]^*$ through the following angles: $2\varphi[\bar{1}10]$, $2\varphi[\bar{3}10]$, $180° = 2\varphi[\bar{1}00]$, $2\varphi[\bar{3}\bar{1}0]$, and $2\varphi[\bar{1}\bar{1}0]$, where $\varphi[uvw]$ is defined as the counter-clockwise angle $[010]:[uvw]$. The angle $\varphi[\bar{3}10]$ is found to be $60° + \varepsilon$, with $\varepsilon = 1'$ from single-crystal data and $\varepsilon = 10''$ from refined powder data. Likewise, $\varphi[\bar{1}10] = 30° + \varepsilon$, $\varphi[\bar{1}00] = 90°$, $\varphi[\bar{3}\bar{1}0] = 120° - \varepsilon$, $\varphi[\bar{1}\bar{1}0] = 150° - \varepsilon$. As can be shown by Euler's theorem, these unorthodox twin operations, each premultiplied by a 180° rotation around the symmetry axis $b[010]$, are *rigorously*

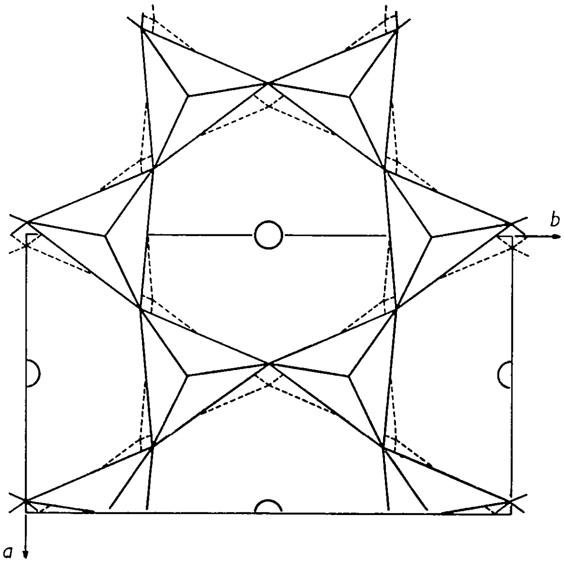

Fig. 1. Orthogonal projection of tetrahedral sheets onto (001). Open circles are potassium ions; dashed lines represent the top tetrahedral sheet in the cell below. Note ditrigonal symmetry of six-membered tetrahedral ring.

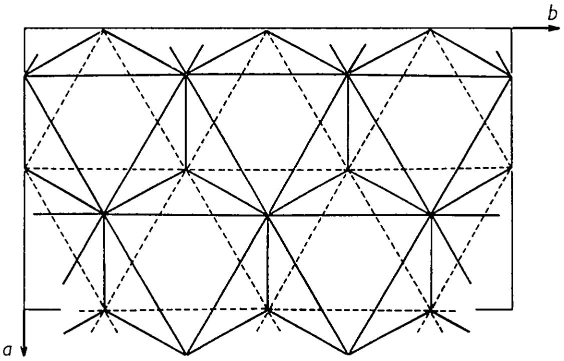

Fig. 2. Orthogonal projection of the octahedral sheet onto (001). Note pseudo 3-fold axes through Fe^{2+} octahedra.

equivalent to 180° rotations around [1$\bar{1}$0], [3$\bar{1}$0], [100], [310], [110], respectively. Sadanaga & Takéuchi (1961) postulate the strict equivalence of the latter (twin rotations) with rotations around c^* through 60°, 120°, 180°, 240°, 300°. As shown above, the angles other than 180° differ from the stated values by 2ε.

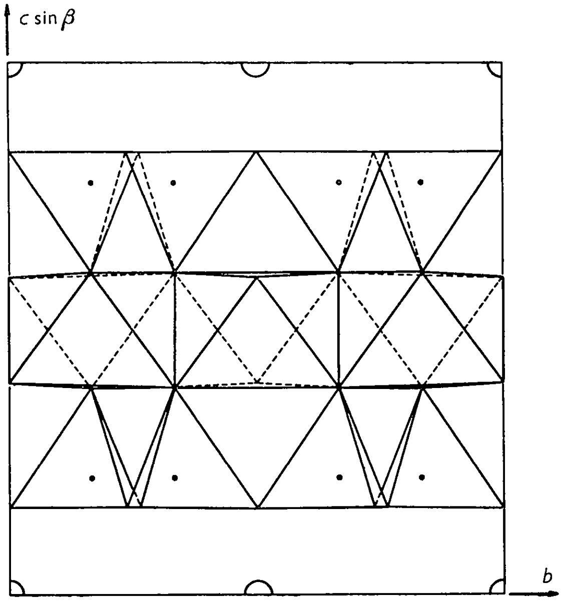

Fig. 3. Orthogonal projection of complete cell onto plane perpendicular to a axis. Open circles are potassium ions; small solid circles, (Si, Fe^{3+}) ions. Note flattening of octahedra into trigonal antiprisms.

The equivalence of the 180° rotation around a twin axis lying in (001) with a rotation around c^* through a multiple of 60° is the key to the mechanism of twinning, which thus turns out to be similar to that postulated by Smith & Yoder (1956) to explain the formation of mica polymorphs.

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