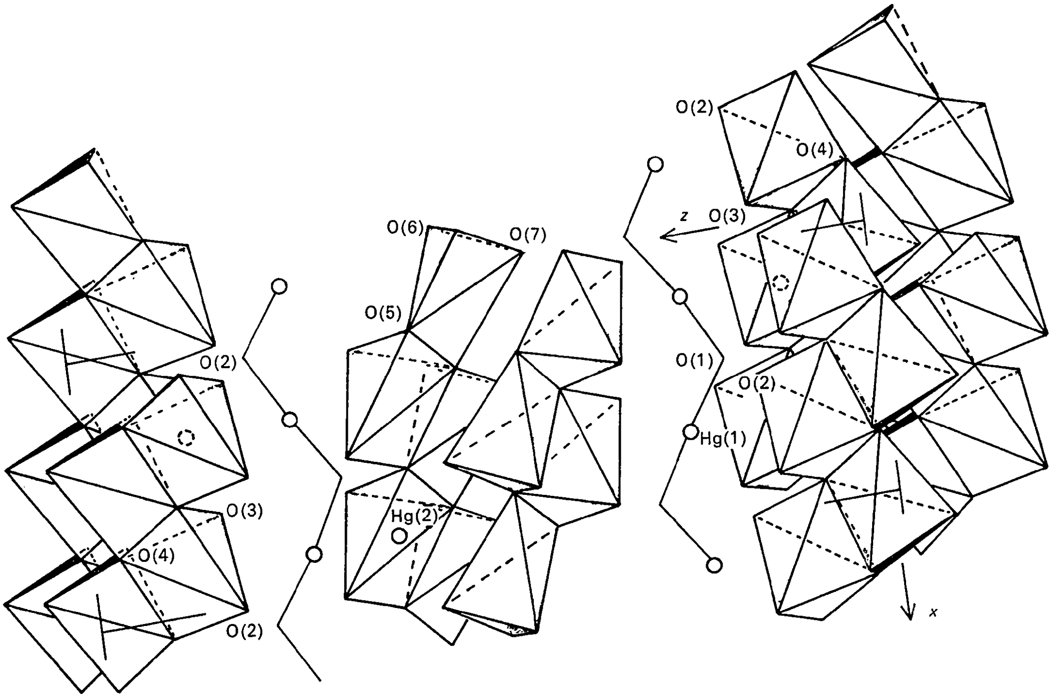

Fig. 8. Vue perspective de la structure suivant y.

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Crystal Structure of a $2M_2$ Lepidolite

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(*Received* 24 *October* 1972; *accepted* 4 *December* 1972)

The crystal structure of a natural sample of a $2M_2$ lepidolite from the Island of Elba, Tuscany (Italy), has been determined and refined by least-squares methods using three-dimensional data collected by the Weissenberg method with Cu $K\alpha$ radiation. This specimen has a composition close to $Pl_{50}Tl_{50}$ (where Pl = polylithionite, Tl = trilithionite) and cell dimensions: $a = 9{\cdot}04 \pm 0{\cdot}02$, $b = 5{\cdot}22 \pm 0{\cdot}02$, $c = 20{\cdot}210 \pm 0{\cdot}001$ Å, $\beta = 99°35' \pm 20'$. The main features of the structure are (*a*) a remarkable octahedral ordering, with one of the octahedral sites filled almost exclusively by Li atoms; (*b*) the tetrahedral sheets, made up by tetrahedra distorted into a form near to an elongated trigonal pyramid, deviate little from hexagonality ($\alpha = 6°27'$).

Introduction

The polymorphism of micas has long been, and still is, as ubject of great interest to mineralogists ever since Hendricks & Jefferson (1939) showed that these minerals crystallize with one of several layered structures, all of which are based on the same substructure. Many studies have been devoted to the problem of enumerating all possible stacking sequences of mica polymorphs, to the experimental methods of identification of such polymorphs, and to the survey of their frequencies of occurrence.

The present trend of research in the field of mica polymorphism is toward a more detailed knowledge of the structure of several polymorphs as an indispensable basis for understanding the factors which control their crystallization.

Following this trend we decided to undertake the determination of the structure of a $2M_2$ lepidolite, whose features should be very important for the evaluation of the structural controls over mica polymorphism. The crystal structure of a $2M_2$ lepidolite was published by Takeda, Haga & Sadanaga (1971) when the present determination was completed and the refinement was in process. As the chemical composition of this $2M_2$ lepidolite was substantially different from that analysed in the present paper, a useful and interesting comparison can be made.

Experimental

A single crystal of a $2M_2$ lepidolite suitable for structural studies has been obtained from the sample, from Elba (Italy), fully described in a previous work by two of the authors. The sample also contains 1M crystals for which, as well as for the $2M_2$ crystals, detailed optical and X-ray data were given (Franzini & Sartori, 1969).

The intensity data were collected with nickel-filtered Cu $K\alpha$ radiation ($\lambda = 1{\cdot}5418$ Å) by means of Weissenberg photographs, using the multiple-film technique and integration process. A transparent colourless flake, plate parallel to (001), with a perfect hexagonal outline up to 1·28 mm across and 0·04 mm in thickness, was carefully chosen from among the least deformed crystals (its Weissenberg photographs showed only minor diffuse scattering and spot broadening). Five layers with b as rotation axis ($k=0$ to 4) were recorded; 525 independent reflexions were observed. The intensities, measured with a Nonius microdensitometer, have been corrected for Lorentz and polarization factors. The absorption correction was made by computing the transmission factor by means of a program (Alberti, 1968) based on the Monte Carlo method proposed by Alberti & Gottardi (1966) for crystals of any shape and absorption: with a linear absorption coefficient of 115·31 cm^{-1} for Cu $K\alpha$ radiation, the transmission factors were found to vary to a great extent, ranging from 0·10 to 0·65.

Determination and refinement of the structure

A hypothetical structure for the $2M_2$ polymorph was proposed by two of us (Franzini & Sartori, 1969) in a previous paper concerned with the crystal data of the Elba lepidolites. According to this hypothesis, based mainly on one-dimensional data, the $2M_2$ polymorph would be built up by the stacking of two mica layers, labelled A and B, differing structurally by the opposite rotations of tetrahedra (deviations from hexagonal symmetry); this structural model would retain an octahedral coordination around the potassium ion, notwithstanding the ditrigonal surface symmetry, for the polymorphs derived from the stacking of mica layers rotated by an odd multiple of 60° (*i.e.* $2M_2$, 2O and 6H).

This hypothesis has not been substantiated by the present study based on three-dimensional data; the full structural analysis, as pointed out also by Takeda *et al.* (1971), has shown that the structural features of the lepidolites cannot be elucidated by means of a one-dimensional Fourier synthesis and that the tendency of the potassium ion to assume an octahedral coordination had been overestimated.

The trial structure was first computed on the basis of such a model of alternating A and B mica layers; the atomic coordinates were derived from those of the basic mica unit (the 1M form), predicted from the cell dimensions and composition (Donnay, Donnay & Takeda, 1964), taking into account the geometric relationships between the A and B layers pointed out by Franzini (1969). The space group chosen as the most probable for this model was $C\bar{1}$. Statistical distribution of aluminum and lithium over all octahedral positions was assumed.

The atomic scattering factors were taken from *International Tables for X-ray Crystallography* (1962), all the atoms being considered in their neutral state. Three cycles of full-matrix least-squares refinement of positional parameters, scale factors and isotropic temperature factors were computed, by means of the program by Busing, Martin & Levy adapted for the Crystal Structures Calculations System X-ray 63 (Stewart, 1964); the resulting unweighted R_1 value (defined as $R_1 = \sum||F_o| - |F_c||/\sum|F_o|$) for the observed reflexions was 0·142. No further improvement was obtained with subsequent refinement cycles, during which the shifts of positional parameters remained smaller than the standard deviations.

At this stage a refinement in the $C2/c$ space group of half of the atoms refined in $C\bar{1}$ was tried; it proved to be successful, since the R_1 value rapidly dropped to 0·130. All the subsequent refinement cycles were then carried out in space group $C2/c$ and the structural model based on alternating A and B mica layers was rejected.

A preliminary computation of the bond lengths showed that the two crystallographically independent octahedra had significantly different mean bond lengths (M_1–O = 1·988, M_2–O = 2·119 Å); this suggested a remarkable ordering in octahedral cation distribution. On the basis of the values given by Shannon & Prewitt (1969) for Li–O (2·12 Å) and Al–O (1·91 Å) distances for lithium and aluminum in octahedral coordination, we computed occupancies corresponding to $Al_{0\cdot63}Li_{0\cdot37}$ for position 8(f) and $Li_{0\cdot95}Al_{0\cdot05}$ for position 4(c). (Actually the M_2–O mean distance did not indicate any isomorphic replacement so that at first only Li was placed in this octahedron; the negative value assumed by its temperature factor forced us to admit for this position a small amount of substitution of aluminum for lithium).

After these changes and two further cycles of refinement the R_1 value was reduced to 0·117. By a careful survey of the calculated structure factors a few mistakes in estimating intensities and indexing reflexions were corrected; the final value assumed by the unweighted R_1 index, for all the 525 observed reflexions, was 0·096, whereas the weighted R_2 index (defined as $[\sum w||F_o|-|F_c||^2/\sum w|F_o|^2]^{1/2}$) reached 0·106 (in all refinement cycles unit weights were assigned to all reflexions).

No further changes of the octahedral occupancies have been made in the last cycles, since the shifts of the M_{oct}–O mean bond lengths were smaller than their standard deviations; hence the final structural formula is the following:

$(K_{0\cdot88}Na_{0\cdot06}Rb_{0\cdot05}Ca_{0\cdot01})\ (Al_{0\cdot63}Li_{0\cdot37})_2\ (Li_{0\cdot95}Al_{0\cdot05})\ (Si_{3\cdot36}Al_{0\cdot64})\ O_{10}\ (F_{1\cdot53}(OH)_{0\cdot47})$.

The observed and calculated factors are compared in Table 1; Table 2 gives the final positional and thermal parameters with their standard deviations.

Table 1. *Observed and calculated structure factors for* 2M$_2$ *lepidolite*

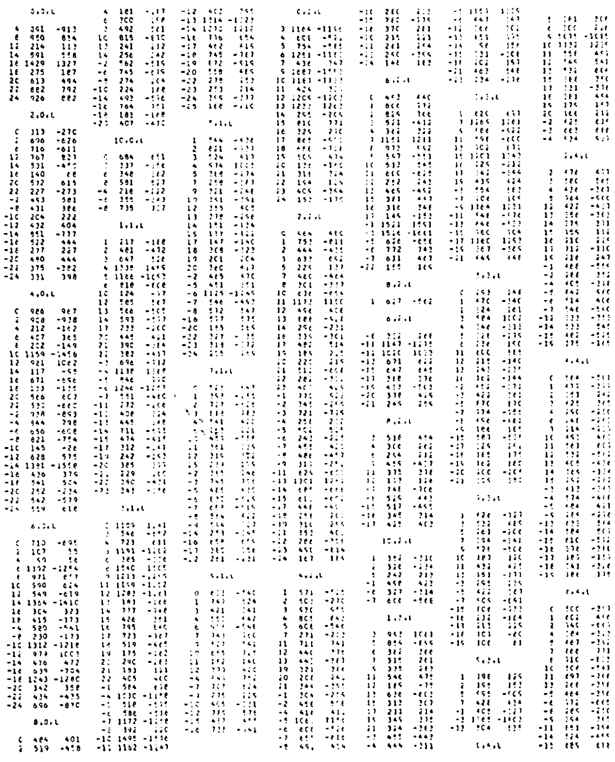

Table 2. *Atomic coordinates (*× 10^4*) and isotropic temperature factors of* 2M$_2$ *lepidolite*

	x	y	z	B(Å^2)
T$_1$	2937 (4)	935 (10)	1338 (2)	0·47 (7)
T$_2$	1251 (4)	5865 (10)	1338 (2)	0·37 (7)
M$_1$	857 (6)	2583 (15)	3 (3)	0·41 (9)
M$_2$	2500	7500	0	0·74 (15)
K	0	916 (11)	2500	1·06 (8)
O$_1$	2675 (10)	1055 (26)	534 (5)	1·12 (17)
O$_2$	908 (10)	5730 (25)	530 (4)	0·93 (16)
O$_3$*	4450 (10)	5743 (26)	497 (4)	1·77 (16)
O$_4$	2070 (12)	3243 (24)	1663 (5)	1·39 (21)
O$_5$	2366 (13)	8217 (25)	1616 (5)	1·55 (22)
O$_6$	4722 (11)	1241 (26)	1655 (5)	1·24 (18)

* O$_3$ stands for (OH+F).

Discussion of the structure

Before discussing in detail the features of the structure, a few remarks about the model proposed by Franzini & Sartori (1969) in their previous paper on Elba lepidolites seem appropriate. On the basis of an α value of about 11°, calculated by assuming the absolute regularity of the tetrahedra, the authors proposed a structure built up by the stacking of A and B mica layers; as a reason for the different features of these layers, they supposed (Franzini, 1969; Franzini & Sartori, 1969) that such mica layers would have different octahedral compositions, namely a composition close to $(LiAl_2)$ for the A layer and a composition close to (Li_2Al) for the B layer. However, the structural analysis did not support any difference in composition between the two octahedral sheets of the cell. In the subsequent refinement carried out in the $C2/c$ space group, which proved to be the correct one, the two sheets were to be considered equivalent; they were shown to be made up of two small octahedra $(Al_{0\cdot63}\ Li_{0\cdot37})$ and a large one $(Li_{0\cdot95}\ Al_{0\cdot05})$. The condition that the mica layers both have an A structure is then realized and the coordination polyhedron around K^+ becomes a trigonal prism. This kind of coordination, as well as the hexagonal prismatic one shown by polylithionite (Takeda & Burnham, 1969), is quite unusual for potassium in the structure of the micas; however, it would be (according to Takeda *et al.*, 1971) a no more unstable configuration when considerable amounts of hydroxyl are replaced by fluorine and when the charge of the basal oxygens is very low, *i.e.* when little Si is replaced by Al in the tetrahedra; this is what generally happens in most of the natural lepidolites. These structural features are associated with a very low tetrahedral rotation angle α (6°27′) compared with the value calculated (10°58′) on the basis of the Radoslovich (1961) formula: $\alpha = \arccos\,(b_0/b_{tetr})$. The low value of α is realized through a considerable distortion of the tetrahedra, which look like elongated trigonal pyramids, with O_{base}–O_{base} = 2·627 Å and O_{base}–O_{apex} = 2·688 Å. This feature, which cannot be predicted using chemical composition and cell dimensions, and the overestimated tendency of potassium to assume an octahedral coordination induced some of us, as we pointed out earlier, to propose a hypothesis which was not later supported by the experimental data obtained by the three-dimensional structural analysis.

From Fig. 1 and Tables 3 and 4, in which the more interesting interatomic distances and angles are reported, the fundamental features of the structure of this polymorph can be deduced.

In the tetrahedral sheet there is no evidence of any cation ordering, since the two crystallographically independent tetrahedra are identical within the precision of the determination. The tetrahedra are rather small in comparison with those of all the other micas, both

Table 3 *Interatomic distances in* $2M_2$ *lepidolite*

T_1 tetrahedron		T_2 tetrahedron	
T_1–O_4	1·634 (13) Å	T_2–O_4	1·640 (13) Å
T_1–O_6	1·642 (11)	T_2–O_5	1·628 (14)
T_1–O_5^{i}‡	1·640 (15)	T_2–O_6^{ii}	1·629 (12)
T_1–O_1*	1·604 (10)	T_2–O_2*	1·612 (10)
Mean	1·630 (6)	Mean	1·627 (6)
O_4–O_6	2·618 (17)	O_4–O_5	2·614 (21)
O_6–O_5^{i}	2·642 (18)	O_5–O_6^{ii}	2·616 (17)
O_5^{i}–O_4	2·640 (21)	O_6^{ii}–O_4	2·635 (17)
O_6–O_1*	2·678 (16)	O_4–O_2*	2·689 (15)
O_5^{i}–O_1*	2·694 (16)	O_5–O_2*	2·698 (16)
O_4–O_1*	2·688 (16)	O_6^{ii}–O_2*	2·684 (15)
Mean	2·660 (7)	Mean	2·656 (7)

M_1 octahedron		M_2 octahedron	
M_1^{iv}–O_1	1·981 (13) Å	M_2–O_2	2·141 (12) Å
M_1^{iv}–O_2^{iii}	1·977 (13)	M_2–O_1^{vi}	2·139 (14)
M_1^{iv}–O_1^{iv}	1·975 (13)	M_2–O_3	2·090 (11)
M_1^{iv}–O_2^{iv}	1·954 (15)	Mean	2·123 (7)
M_1^{iv}–O_3†	2·005 (16)		
M_1^{iv}–O_3^{v}	1·990 (13)		
Mean	1·980 (6)		

Unshared		Unshared	
O_1–O_2^{iii}	2·929 (15)	O_3–O_2	3·214 (15)
O_1–O_3	2·934 (20)	O_1^{vi}–O_3	3·211 (20)
O_3–O_2^{iii}	2·926 (20)	O_2–O_1^{vi}	3·205 (20)
O_1^{iv}–O_2^{iv}	2·916 (20)	Mean	3·210 (11)
O_1^{iv}–O_3^{v}	2·908 (15)		
O_3^{v}–O_2^{iv}	2·914 (20)		
Mean	2·921 (7)		

Shared	
O_2^{vii}–O_3	2·753 (17) Å
O_1^{iv}–O_3	2·753 (16)
O_1^{iv}–O_2	2·837 (16)
Mean	2·781 (9)
O_3—O_3^{v}	2·508 (15)

Interlayer cation

Inner		Outer	
K^{viii}–O_6	2·965 (16) Å	K^{viii}–O_4	3·214 (15) Å
K^{viii}–O_5	2·981 (15)	K^{viii}–O_6^{vi}	3·250 (17)
K^{viii}–O_4^{viii}	2·982 (14)	K^{viii}–O_5^{iii}	3·320 (15)
Mean	2·976 (9)	Mean	3·261 (9)
	K^{viii}–O_3	3·997 (10) Å	

Shortest interlayer distances		
O_6—O_6^{ix}	3·368 (14) Å	(×2)
O_4^{viii}–O_5^{ix}	3·430 (15)	(×4)
Mean	3·409 (8)	

* Apical oxygens

† O_3 Hydroxyl and fluorine

‡ The atoms of the different asymmetric units are related to the symmetry-equivalent atoms of the fundamental unit as follows:

i	$x,\ y-1,\ z$	v	$1-x,\ 1-y,\ -z$
ii	$x-\frac{1}{2},\ y+\frac{1}{2},\ z$	vi	$x,\ 1+y,\ z$
iii	$x+\frac{1}{2},\ y-\frac{1}{2},\ z$	vii	$\frac{1}{2}-x,\ \frac{3}{2}-y,\ -z$
iv	$\frac{1}{2}-x,\ \frac{1}{2}-y,\ -z$	viii	$\frac{1}{2}+x,\ \frac{1}{2}+y,\ z$
		ix	$1-x,\ y,\ \frac{1}{2}-z$

Table 4. *Interatomic angles in* $2M_2$ *lepidolite*
Central atom is the vertex

T_1 tetrahedron		T_2 tetrahedron	
O_5^{i}–T_1–O_6†	107·20 (64)°	O_6^{ii}–T_2–O_5	106·89 (67)°
O_5^{i}–T_1–O_4	107·50 (68)	O_6^{ii}–T_2–O_4	107·40 (65)
O_6–T_1–O_4	106·09 (63)	O_5–T_2–O_4	106·19 (65)
O_1^*–T_1–O_6	111·18 (63)	O_2^*–T_2–O_4	111·53 (66)
O_1^*–T_1–O_4	112·23 (64)	O_2^*–T_2–O_5	112·71 (66)
O_1^*–T_1–O_5^{i}	112·29 (66)	O_2^*–T_2–O_6^{ii}	111·77 (58)
Mean	109·42 (26)	Mean	109·42 (26)

Angles between basal oxygens:		Amount of rotation from 120°
O_6—O_4—O_5	107·28 (57)°	12·72°
O_4—O_5—O_6^{vi}	132·80 (60)	12·80
O_5—O_6^{vi}—O_4^{viii}	106·85 (66)	13·15
O_6^{vi}—O_4^{viii}–O_5^{iii}	132·55 (59)	12·55
O_4^{viii}–O_5^{iii}–O_6	106·98 (58)	13·02
O_5^{iii}–O_6—O_4	133·23 (70)	13·23
	Mean	12·91 (25)

Tetrahedral rotation angle $\alpha = 6{\cdot}45\ (13)°$.

* Apical oxygens.

† The atoms of the different asymmetric units are related to the symmetry equivalent atoms of the fundamental unit as follows:

i	$x,\ y-1,\ z$	v	$1-x,\ 1-y,\ -z$
ii	$x-\frac{1}{2},\ y+\frac{1}{2},\ z$	vi	$x,\ 1+y,\ z$
iii	$x+\frac{1}{2},\ y-\frac{1}{2},\ z$	vii	$\frac{1}{2}-x,\ \frac{3}{2}-y,\ -z$
iv	$\frac{1}{2}-x,\ \frac{1}{2}-y,\ -z$	viii	$\frac{1}{2}+x,\ \frac{1}{2}+y,\ z$
		ix	$1-x,\ y,\ \frac{1}{2}-z$

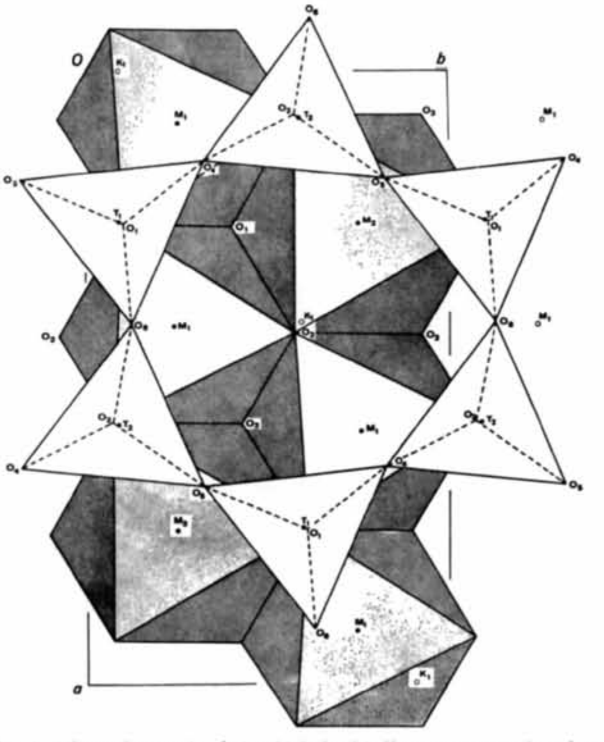

Fig. 1. View along **c*** of the $2M_2$ lepidolite structure showing one $(Si_2O_5)^{2-}$ ring and part of an octahedral sheet.

dioctahedral and trioctahedral, because of the little replacement of Al for Si. Moreover, as we have already emphasized, these coordination polyhedra are remarkably distorted into a form near to the trigonal pyramid.

Noteworthy is the shortening of the $Si–O_{apex}$ distance (mean length 1·608 Å), compared with the $Si–O_{base}$ distance (mean length 1·635 Å), a peculiarity which is also encountered in the $2M_2$ lepidolite structure determined by Takeda *et al.* (1971), and it is even more evident in the polylithionite structure. This behaviour can be explained on the basis of the *d–p* π bond theory suggested by Cruickshank (1961) as well as on the basis of the Pauling–Zachariasen method of the balancing of valences. If appreciable π bonding is responsible for the short $Si–O_{apex}$ bond, the bond angles subtended at silicon by the tetrahedral edges and involving O_{apex} should be affected. They are indeed, with angles 111 to 113°, whereas the angles subtended by the edges made of bridging oxygen atoms range from 106 to 107·5° (Table 4).

In the octahedral sheet we find a considerable ordering of the cations; in fact the lithium ions almost completely fill the 4(*c*) position before entering the 8(*f*) position. Consequently the octahedral sheet appears to be built up from a large octahedron and two smaller ones, assuming a configuration rather similar to that of the corresponding sheet of the dioctahedral micas, with the difference that whereas the Li-rich octahedron is rather close in size to an empty octahedron, the other two octahedra, which still contain an appreciable amount of lithium ($Al_{0·63}Li_{0·37}$), are larger than the filled octahedra of the dioctahedral micas. As a result the lateral dimensions of the octahedral sheet are rather large, also owing to the flattening of such coordination polyhedra (mainly of the Li-rich octahedron, $\psi = 60°47'$).

It is then evident that the misfit between such an octahedral sheet and a tetrahedral sheet characterized by small elongated tetrahedra is quite small, and consequently the tetrahedral rotation needed for the adjustment of one sheet to the other is also small. The 'tetrahedral tilt' (calculated according to the formula: $\Delta z = [(ZO_6 + ZO_4)/2 - ZO_5]\ .\ c \sin\beta)$ is also small; the value $\Delta z = 0·086$ attests that the O_{basal} sheet is, on the whole, little corrugated.

As in other micas the K–O distances are of two kinds, due to the reduced, but always manifest, ditrigonal configuration of the oxygen rings on either side of the potassium ion. The observed values (Table 3) give averages of 2·976 and 3·261 Å for the inner and the outer K–O distances, respectively. Since the K–O distances depend on the size of the tetrahedra and on their rotation angle (Takeda & Burnham, 1966), the small size of the tetrahedra and the low value of α account for the small value of the longer of the two K–O distances and explain why the shorter K–O distance is as large as it is. Since in $2M_2$ polymorph the two mica layers are rotated by 60° on each other, the coordination polyhedron around K^+ is then a trigonal prism if we take into account only the 6 inner oxygens, a ditrigonal prism if we consider also the 6 outer oxygens.

A detailed comparison of the parameters of our mica with those of the other two lepidolites whose structures are reported in the literature now seems appropriate. Most interesting, in our opinion, is a comparison with Takeda's $2M_2$ lepidolite structure. Tables 5 and 6 show in fact a close analogy between the structure of the Elba lepidolite and that of the Rozna lepidolite. However, these crystals are substantially different in composition; if we express their composition in terms of the three end members, polylithionite (Pl) [$KLi_2AlSi_4O_{10}F_2$], trilithionite (Tl) [$KLi_{3/2}Al_{3/2}Si_3AlO_{10}(F,OH)_2$] and muscovite (Ms) [$KAl_2Si_3AlO_{10}(OH)_2$], the composition of our lepidolite is close to $Pl_{50}Tl_{50}$, whereas that of the Takeda's mica is $Ms_{40}Pl_{30}Tl_{30}$. These compositions, plotted in the ternary diagram of Munoz (1968), turn out to be of great interest, since Takeda's mica is very close to the region of the mixed-layer Li-micas, while our mica lies on the Pl–Tl join, in the field of the 1M and $2M_2$ lepidolites.

Table 5. *Cell dimensions and structural formulae of* $2M_2$ *lepidolites*

	Lepidolite, $2M_2$*	Lepidolite, $2M_2$†
a	9·04 (2) Å	9·032 (2) Å
b	5·22 (2)	5·200 (3)
c	20·210 (1)	20·15 (4)
β	99° 35′ (20′)	99° 46′ (10′)
Space group	*C*2/*c*	*C*2/*c*

* $(K_{0·88}Na_{0·06}Rb_{0·05}Ca_{0·01})(Al_{0·63}Li_{0·37})_2(Li_{0·95}Al_{0·05})(Si_{3·36}Al_{0·64})O_{10}(F_{1·53}(OH)_{0·47})$: Elba, Italy. Present work.

† $(K_{0·87}Na_{0·12}Rb_{0·06}Ca_{0·02})(Al_{0·65}Li_{0·35})_2[Li_{0·35}Al_{0·10}(Fe^{2+}, Mn, Mg)_{0·15}\square_{0·40}](Si_{3·39}Al_{0·61})O_{10}(F_{1·2}(OH)_{0·8})$: Rozna, Moravia, Czechoslovakia. Takeda *et al.* (1971).

Table 6. *Comparison of some interatomic distances in* $2M_2$ *lepidolites*

	$2M_2$ lepidolite from Elba (Present work)	$2M_2$ lepidolite from Rozna, Moravia (Takeda *et al.*, 1971)
	Tetrahedral sheet	
Mean T_1–O	1·630 (6) Å	1·622 (4) Å
Mean T_2–O	1·627 (6)	1·633 (4)
	Octahedral sheet	
Mean M_1–O	1·980 (6) Å	1·967 (3) Å
Mean M_2–O	2·123 (7)	2·144 (4)
	Interlayer region	
Mean K–O (inner)	2·976 (9) Å	2·980 (5) Å
Mean K–O (outer)	3·261 (9)	3·220 (5)

These compositions are calculated on the basis of chemical analyses of samples which contain both the 1M and $2M_2$ polymorphs. The doubt arises that such compositions are not necessarily related to the $2M_2$ polymorphs structurally analysed and that the close resemblance between the structural parameters reported by Takeda *et al.* and ours is to be ascribed to the true chemical compositions of the crystals being much closer to each other than the bulk compositions of the samples. Though lacking chemical data on a single crystal, we believe that the satisfactory refinements of the structures rule out this possibility; the reason for the close resemblance of the two structures is then to be found in other features. We think such a reason most likely lies in the following two peculiarities.

The two $2M_2$ lepidolites have tetrahedral sheets nearly equal in composition ($Si_{3\cdot39}Al_{0\cdot61}$ = Rozna lepidolite; $Si_{3\cdot36}Al_{0\cdot64}$ = Elba lepidolite) and so are very close in dimensions.

They have octahedral sheets which, though clearly different in composition, are both made up of a large octahedron and two smaller ones, whose sizes are very close in the two lepidolites. Such a close geometric resemblance of the octahedral sheets of the two crystals is achieved through an almost equal composition of the small octahedra ($Al_{0\cdot65}Li_{0\cdot35}$ = Rozna lepidolite; $Al_{0\cdot63}Li_{0\cdot37}$ = Elba lepidolite) and through the concentration in the larger octahedron of the Rozna lepidolite of all the vacancies, while the corresponding site of the Elba lepidolite accomodates almost only lithium.

The 1M polylithionite studied by Takeda & Burnham (1969), in spite of its different polymorphic type, has tetrahedral and octahedral sheets which show features rather close to those of the $2M_2$ lepidolites. They are even more evident because of the presence of only Si in the first sheet and of the higher content of Li of the second; as a consequence the misfit between them is even less and the tetrahedral rotation angle α lowest ($\alpha = 3°$).

To sum up, we can say that the most interesting feature which emerges from the comparison between all the structurally analysed lepidolites is the constant presence of remarkable octahedral ordering. The Li–Al substitution is characterized by concentrating in an octahedral site almost exclusively Li (0·90 to 0·95 in polylithionite and in the Elba lepidolite) or Li plus vacancies (0·75 in the Rozna lepidolite), and by filling the other two sites with Al plus the remaining Li. It is in this distorted octahedral sheet and in its consequently easy adaptability to the tetrahedral sheet without a great deviation from hexagonality of the latter, that we think lies the main difference between the structure of the polylithionite-rich lepidolites and that of the lepidolites close to trilithionite, characterized by having the $2M_1$ as the more stable polymorph (Munoz, 1968). The discontinuity observed by Munoz (1968) between them would then be a discontinuity which separates a structure with a remarkable octahedral ordering from one with a disordered octahedral sheet. Rieder (1968), in his paper on lithium-iron micas, also suggested that the octahedral sheet of the trilithionites must be disordered.

However the final answer to the problem of the relations between the structure of trilithionites and that of the other lepidolites and between the former and that of the $2M_1$ lithian muscovites can be given only by the crystal structure analysis of a $2M_1$ trilithionite.

This work was supported by Consiglio Nazionale delle Ricerche, Roma.

All the calculations were performed on the IBM 7090 computer of the Centro Nazionale Universitario di Calcolo Elettronico (Pisa).

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