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Complex stacking sequences in a lepidolite from Tørdal, Norway

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ABSTRACT

Lepidolite from Tørdal, Norway, has been described as the complex stacking polytype $3M_2 = [1\bar{2}1]$ of space group $C2$. Subsequent optical second-harmonic analysis indicated a center of symmetry possibly present, and Weissenberg photographs showed weak, extra six-layer spots. Of the 402 possible six-layer mica structures, only eight have the observed monoclinic symmetry and unit-cell geometry. Stacking arrangement $6M_1 = [1\bar{2}1\bar{1}2\bar{1}]$ of space group $C2/c$, which can be generated by adding a center of symmetry in the octahedral sheet of the $3M_2$ structure, fits the observed intensity-data best. The $6M_1$ structure would refine only to $R = 23\%$, however, and anomalies on DED maps in sections where the $3M_2$ and $6M_1$ structures differ suggest that the $3M_2$ structure is dominant. Refinement of the data as a $3M_2$ structure proceeded better to $R = 12\%$, but is incorrect in detail.

HRTEM imaging showed that both three-layer and six-layer stacking sequences are present in the same crystal, that twins formed by stacking faults of the three-layer structure exist, and that material with substantial stacking disorder also exists.

INTRODUCTION

Micas crystallizing as complex polytypes—stacking arrangements characterized by having several different interlayer rotation angles between layers—are rare. The $3M_2$ stacking arrangement is the most frequently identified complex polytype reported in the literature. Lepidolites at several localities are reported to have this structure (Heinrich et al., 1953; Neumann et al., 1957; Christie, 1961; Bailey and Christie, 1978). The present study involves a more detailed investigation of the crystal structure of one of these lepidolites.

Christie (1961) first reported the occurrence of a two-layer orthorhombic lepidolite in a pegmatite at Tørdal, Norway. Bailey and Christie (1978) re-examined the material and described it as polytype $3M_2$ with ideal space group $C2$. The formula unit given by these authors is $(K_{0.87}Na_{0.03})(Al_{1.31}Li_{1.08}\ Mn_{0.27}Fe_{0.08})\ (Si_{3.37}Al_{0.63})O_{10}$-$(F_{1.19}OH_{0.81})$. A subsequent optical second-harmonic analysis of the sample (Horsey, 1981) gave no signal, thus indicating that the structure possibly has a center of symmetry. This finding contradicted the noncentrosymmetric symmetry of space group $C2$. Because of this discrepancy, further research to resolve the true nature of the layer stacking sequence was undertaken. Weissenberg photographs revealed the presence of weak "extra" reflections denoting a six-layer periodicity along Z. Thus, the sample either represented a six-layer mica, a twinned three-layer arrangement, or a submicroscopic intergrowth of three-layer and six-layer structures. No macroscopic twinning was visible under the polarizing microscope.

The purposes of this study were to (1) determine the actual polytype present and (2) refine the crystal structure so that the crystal chemistry of this unusual lepidolite could be assessed. The first step in this investigation was to treat the mica as a six-layer structure and determine its stacking arrangement.

All specimens used in this study are on repository with the Department of Geology and Geophysics Museum, University of Wisconsin, Madison, under file number UW 1769.

REFINEMENT AS A SIX-LAYER STRUCTURE

It was noted first of all that agreement between observed structure amplitudes (F_o) and those calculated (F_c) from the ideal coordinates for the $3M_2$ structure was excellent for $h0l$ reflections but only moderately good for $0kl$ reflections. This suggested that the true structure would be identical to that of $3M_2$ in [010] projection and could be derived by adding a center of symmetry in the octahedral sheet or interlayer of the $3M_2$ structure so that the observed monoclinic symmetry and cell shape could be preserved. In this manner, four structures can be generated that have the observed cell geometry, but only two of them ($[1\bar{2}1\bar{1}2\bar{1}]$ and $[121\bar{1}\bar{2}\bar{1}]$) have the observed monoclinic symmetry. One ($6M_1$) of these latter two structures eventually proved to be the best fit of all possible six-layer structures, but proof of such best fit required a more systematic survey of all possibilities.

H. Takeda (Univ. Tokyo, pers. comm. to Bailey, 1980) kindly provided a list of the vector stacking symbols for the 402 possible six-layer mica polytypes. The method of describing a stacking sequence of mica layers formulated

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TABLE 1. Cell dimensions of the Tørdal lepidolite

UW Museum no.:	1769/3	1769/3	1769/3	1769/5	1769/4
Radiation:	Mo$K\alpha$	Fe$K\alpha$	Cu$K\alpha$	Cu$K\alpha$	Cu$K\alpha$
a (Å)	5.2372(9)	5.2385(7)	5.2403(8)	5.2382(6)	5.2389(8)
b (Å)	9.071(2)	9.076(1)	9.077(2)	9.074(1)	9.076(2)
c (Å)	59.74(1)	59.74(1)	59.71(1)	59.783(9)	59.76(1)
α (°)	89.97(1)	90.02(2)	90.01(1)	90.04(1)	90.00(2)
β (°)	92.66(1)	92.62(2)	92.61(1)	92.61(1)	92.59(1)
γ (°)	90.00(1)	89.97(1)	89.99(1)	90.01(1)	90.00(2)
V (Å^3)	2835.0(8)	2837.3(9)	2836.7(9)	2838.5(7)	2838.7(9)

by Ross et al. (1966) using vector stacking symbols is employed in this study. Each vector stacking symbol consists of six digits, one for each layer in the unit cell. Each number in the sequence indicates the interlayer stacking angle between that layer and the previous layer and can have the values 0, ±1, ±2, or 3 which refer, respectively, to 0°, ±60°, ±120°, or 180° angles.

Experimental details

Precession and/or Weissenberg X-ray photographs of 45 different crystals of the Tørdal lepidolite sample were taken and examined. All crystals exhibited the weak "extra" six-layer diffraction spots, and a few had twelve-layer spots in addition. Crystals of the latter type were assumed to be twinned and were not used. The three best crystals—those with diffraction spots that exhibited the least amount of streaking and with the sharpest extinction under the polarizing microscope—were chosen for use in this study. The large number (402) of possible six-layer polytypes made it impractical to model and test all of them with the final data. Fortunately, cell geometry and symmetry considerations enabled this number to be reduced systematically to just a few polytypes. The steps by which this was accomplished are explained below.

Cell geometry

Cell dimensions for three different crystals (Table 1) were determined from least-squares refinement of fifteen reflections measured on a Syntex P2$_1$ single-crystal diffractometer using monochromatic Mo, Cu, or Fe/$K\alpha$ X-radiation. The best measurements of cell geometry for this mineral as determined in this study were a = 5.2382(6), b = 9.074(1), c = 59.76(1) Å, and β = 92.61(1)°. This monoclinic-shaped cell eliminates those stacking arrangements resulting in orthorhombic or higher symmetries and those with triclinic-shaped cells.

Symmetry

Intensity data obtained from Weissenberg films and single-crystal diffractometry indicated monoclinic symmetry with $I(hkl) = I(h\bar{k}l)$. No violations of C centering were observed on the zero or upper-level Weissenberg films; therefore, polytypes based on a primitive lattice can be excluded.

No optical second-harmonic signal was detected when the Tørdal lepidolite sample was tested by Horsey (1981), indicating that the crystal structure possibly has a center of symmetry. The $N(z)$ statistical test for a center of symmetry of Howells et al. (1950) was also applied to data collected on the single-crystal diffractometer (hkl reflections with $k = 3n$ were excluded to avoid hypersymmetry) (Rule, 1985). The resulting graph is closest to that of a centrosymmetric structure.

Possible alpha and beta angles

The only polytypes now remaining are the 19 with space group symmetries $C2/c$ and $C2/m$. Possible α and β crystallographic angles were calculated for each of these polytypes. The polytypes formed three groups: one group in which α, not β, was the obtuse angle, a group in which β was ideally 90.00° or 95.01°, and a final group in which β was ideally 91.65° or 93.35°. If the unit cells of polytypes in the first group are put into the standard setting so that β, rather than α, is the obtuse angle with Y the unique axis, then the usual X and Y axes must be reversed. This was not observed in the study specimen, and polytypes in this group can be eliminated. The ideal β angles of stacking arrangements in the second group differ too greatly from the observed β angle of 92.61° for any of these to be the correct polytype. Finally, the eight polytypes in the third group have possible ideal β angles that are very close to the observed β angle of 92.61°. These polytypes were examined in more detail.

Observed beta angle

The eight remaining polytypes have ideal β angles that can be given as either 91.65° or 93.35°. The two different β angles are produced by drawing the resultant Z axis of the stacking sequence to different lattice points. Both resultants and β angles are valid but require that the same reflection be indexed differently for the two cells. It is necessary therefore to determine which of these two angles the observed β represents. Ideal atomic positions were calculated for each of the eight polytypes. The crystallographic least-squares program ORFLS (Busing et al., 1962) was used to calculate structure factors for reflections. Calculated values for each β angle and model were compared with observed structure factors. In each case, the R value for the 93.35° β angle was lower than that calculated for the 91.65° value, indicating that the former is correct.

R values of the eight ideal polytypes

Residuals (R) were calculated for the ideal atomic positions of the eight stacking arrangements based on the β angle of 93.35° (Table 2). F values from 304 uncorrected

TABLE 2. *R* values of ideal six-layer models

Vector stacking sequence	Space group	*R* value (%) $\beta = 93.35°$
$[1\bar{2}1\bar{1}\bar{2}\bar{1}]$	*C2/c*	38.7
$[121\bar{1}\bar{2}\bar{1}]$		44.7
$[011\bar{0}\bar{1}\bar{1}]$		46.9
$[101\bar{1}0\bar{1}]$		48.5
$[233\bar{2}33]$		53.8
[033330]	*C2/m*	63.4
[003300]		80.2
[003030]		>80.0

observed intensities from a short preliminary data collection on a single-crystal diffractometer were used. Stacking arrangement $[1\bar{2}1\bar{1}\bar{2}\bar{1}]$ of space group *C2/c* fits the data best. This is one of the two monoclinic structures predicted originally and is termed the $6M_1$ polytype in this paper.

Refinement of the $6M_1$ polytype

Several attempts were made to refine the $6M_1$ polytype. Data for crystal UW Museum no. 1769/3 were collected first on a Syntex $P2_1$ single-crystal diffractometer using Mo$K\alpha$ radiation (16 708 reflections merged to 2501 independent monoclinic reflections), but it was discovered that the short-wavelength Mo radiation and large *c* cell dimension caused X-ray reflections to overlap. Data were recollected using Cu$K\alpha$ (1257 merged reflections) and Fe$K\alpha$ (1086 merged reflections) radiations in the $2\theta{:}\theta$ variable-scan-rate mode. The integrated intensity (I) was calculated from $I = [S - (B1 + B2)/Br]Tr$, where S is the scan count, B1 and B2 the background counts, Br the ratio of background time to scan time, and Tr the $2\theta{:}\theta$ scan rate in degrees per minute. The integrated intensities were corrected for Lp factors and for absorption by the semi-empirical psi-scan technique of North et al. (1968). After four cycles of least-squares refinement with both the Fe$K\alpha$ and Cu$K\alpha$ data sets, some tetrahedral bond lengths became unrealistically small. A triclinic difference-electron-density map (DED) was prepared at this point so that possible distortions or ordering patterns that violated monoclinic symmetry could be detected. The DED map showed interesting anomalies in the upper tetrahedral sheet of the second layer in sections containing the tetrahedral cations at $z/c = 0.212$, basal oxygens at $z/c = 0.225$, and the interlayer cations at $z/c = 0.250$. In all of these sections and in sections related to these by the *c* glide plane, large anomalies of adjacent positive and negative areas of electron density, sometimes equal to more than 5 e/Å^3, occurred (Fig. 1). Sometimes these anomalies were centered on an atomic position, but often they occurred in vacant areas that would have been occupied if a different stacking angle had occurred in that layer of the polytype. In order to test whether these anomalies were due to the wrong origin being chosen for the stacking sequence, the origin was moved to each of the eight possible unique locations on centers of symmetry and one cycle of least-squares refinement was run. DED maps for each origin were made and examined, and the origin producing a map with the smallest number and magnitude of anomalies was used for refinement. Because the DED maps had also indicated that the tetrahedra should be rotated, basal oxygens were rotated 8° to new positions. Even with these changes, the $6M_1$ polytype would not refine below an unweighted R value of 23.3%. This indicates that this model is incorrect in detail. Refinement of the next-best six-layer polytype from Table 2, $[121\bar{1}\bar{2}\bar{1}]$, was also attempted along with two triclinic polytypes, $[11\bar{2}2\bar{1}\bar{1}]$ and $[0\bar{1}20\bar{2}1]$ (all three of which, like the $6M_1$ polytype, can be produced by adding a center of symmetry somewhere in the $3M_2 = [1\bar{2}1]$ structure). These refinements also failed.

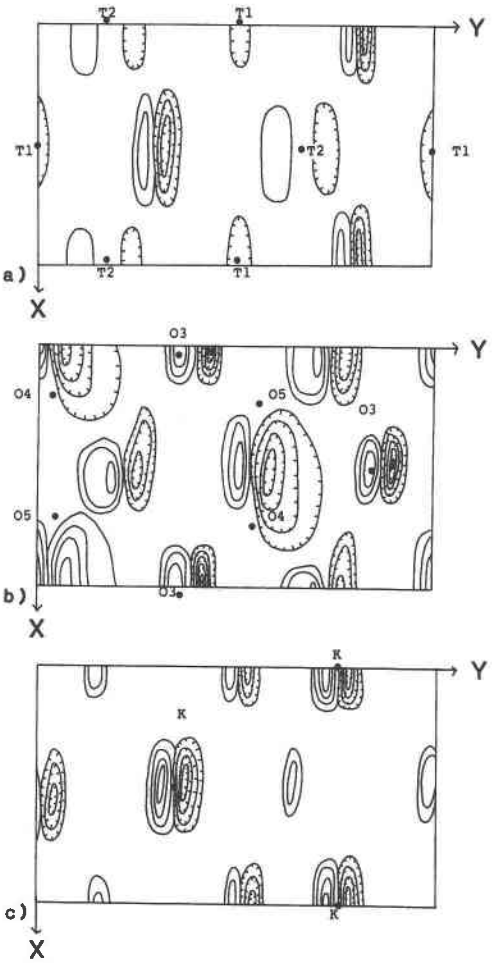

Fig. 1. Anomalies on DED sections along *Z* for $6M_1$ structure. Contour interval = 1 e/Å^3. Elongation of contours is an artifact due to different scales along *X* and *Y*. (a) Tetrahedral cations at $z/c = 0.212$, (b) basal oxygens at $z/c = 0.225$, and (c) K at $z/c = 0.250$.

Periodic intensity distributions

One last search through the 402 possible six-layer polytypes was made in order to be sure that the true polytype

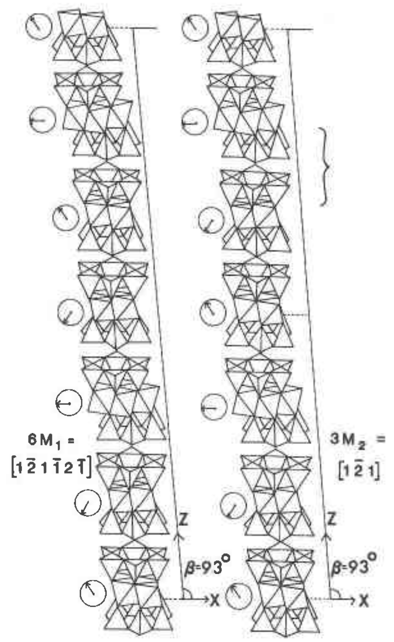

Fig. 2. Comparison of the *6M₁* structure (left) and *3M₂* structure (right). Only *y/b* coordinates for atoms within brackets are different in the two structures. Arrows within circles show plan views of *a*/3 tetrahedral shifts within the layers.

had not been overlooked. M. Ross (U.S. Geol. Survey, pers. comm. to Bailey, 1984) kindly provided computer-generated tables of periodic intensity distributions for certain groups of reflections for each of the 402 polytypes. The data set collected with Cu*Kα* radiation was used to calculate observed periodic intensity distributions. The only polytype that came close to matching the observed pattern of intensities was that of the $6M_1$ polytype. This confirmed that the best six-layer polytype had been chosen for the original refinement. The unmodulated intensities of the "six-layer" spots calculated for this model, however, were very much stronger than those observed experimentally. Therefore, the question arises whether the observed weaker "extra" *hkl*, l = odd reflections are consistent with a true six-layer structure. Because the $6M_1$ polytype would not refine adequately, it must be concluded that the "extra" spots are not generated entirely by a six-layer structure, but by some other mechanism such as twinning of a three-layer structure or by an intergrowth of three-layer and six-layer structures.

REFINEMENT AS A TWINNED THREE-LAYER STRUCTURE

Relationship of the $6M_1$ polytype to the $3M_2$ polytype

If the Tørdal lepidolite is not a six-layer structure, why did the $6M_1$ polytype refine at all? The answer must be that the $6M_1$ and $3M_2$ polytypes are very closely related. The $6M_1$ polytype can be generated from that of the $3M_2$ by adding a center of symmetry in the octahedral sheet, but the similarity between the two structures goes far beyond this. If the ideal positions of the 240 atoms in the $6M_1$ stacking arrangement are compared to the ideal positions of the 240 atoms in two repeat units of the $3M_2$ structure, only 18 of the 240 atoms, or 7.5%, are in different locations. Furthermore, these 18 atoms have the same x/a and z/c coordinates in both structures; they differ only in their positions along *b*.

Figure 2 shows the two stacking arrangements side by side. They are identical in [010] projection. The atoms that are different in the two structures are the interlayer cations at $z/c = 0.75$ and the basal oxygens and some of the tetrahedral cations that surround them. The large anomalies found on DED maps during refinement of the $6M_1$ structure can now be explained. These anomalies centered around cations located at $z/c = 0.25$ and the level related to this by the *c* glide plane, $z/c = 0.75$. The latter level is exactly the area in which atoms of the two structures lie in different positions. In addition, *h0l* reflections showed better agreement between observed and calculated *F* values than *hk0* or *0kl* reflections, as would be expected since both ideal structures only differ in the *Y* direction.

Evidence for twinning

Weissenberg films were re-examined in detail for evidence of twinning. On films from one crystal, the "extra" six-layer–type reflections were long and narrow, a different shape than the circular three-layer spots, indicating that only part of the crystal in the twinned orientation diffracted to produce the six-layer spots. Also, the "extra" six-layer spots on Weissenberg films from several crystals were much more intense than those for other crystals, although three-layer–type reflections for all these crystals had about the same intensities. This implies that the crystals had different amounts of twinning. Finally, in the absence of anomalous dispersion, Friedel's law should hold for an untwinned, noncentrosymmetric crystal with the $3M_2$ structure. There were many sets of reflections in data collected from several different crystals, however, that violated Friedel's law. These violations were determined to occur for reflections that satisfy the geometry required for overlap due to twinning.

Refinement of the $3M_2$ structure

As a preliminary test of the $3M_2$ model, a refinement was attempted using the six-layer data reindexed as a three-layer structure. All of the *hkl* reflections with l = odd were eliminated from the data, and the remaining data were reindexed by dividing l by 2 as if they were collected for a three-layer structure. It should be noted that eliminating the "extra" six-layer spots supposedly caused by twinning does not eliminate all of the possible effects of that twinning. Diffraction spots from the twinned part of the crystal may superimpose over other three-layer–type spots and so affect their intensities. Nevertheless, it was believed that preliminary refinement of the $3M_2$ structure using data reindexed in the manner de-

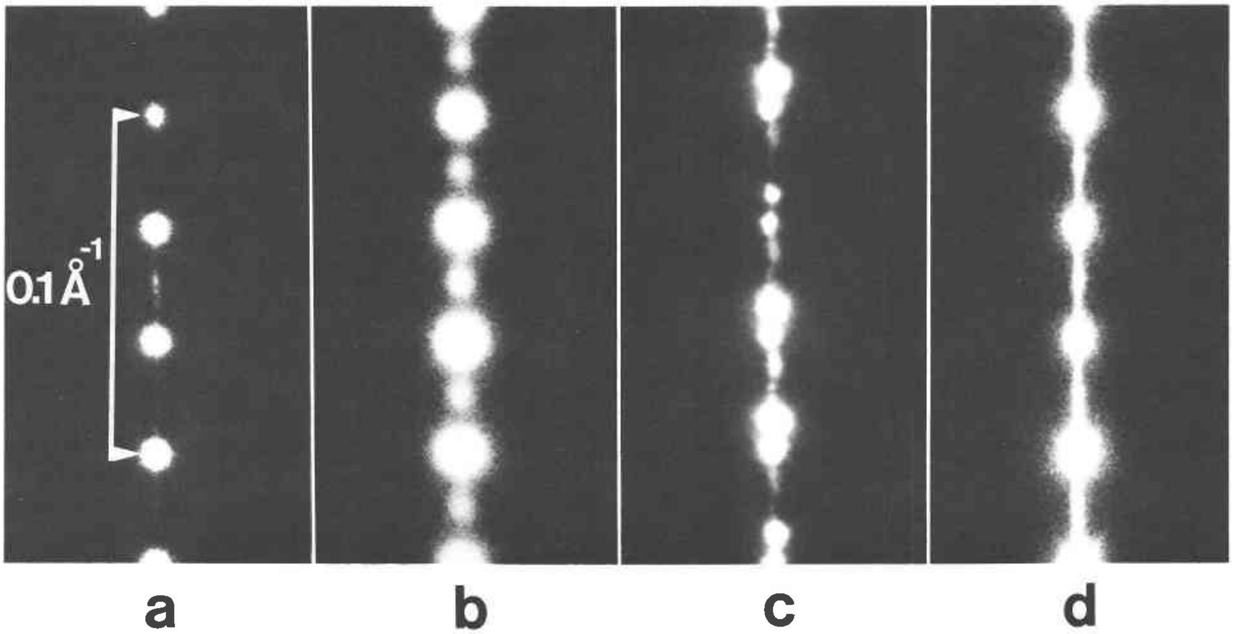

Fig. 3. Portions of the 02*l* rows from selected-area electron-diffraction patterns of Tørdal lepidolite. (a) Pattern from region of predominantly three-layer polytype with some stacking disorder. (b) Pattern from six-layer polytype. (c) Pattern from twinned three-layer structure showing irregular spacing of diffraction spots. (d) Pattern from region with highly disordered stacking.

scribed above might show whether the $3M_2$ structure is indeed basically correct for this lepidolite.

The 32 ideal atomic positions used for this refinement of polytype $[1\bar{2}1] = 3M_2$ are shown in Table 3. The ideal space group of the $3M_2$ structure is $C2$. The origin was chosen in the same place as for the final attempted refinement as a six-layer structure. The position of the interlayer cation (the heaviest atom in the structure) at $z/c = 0.50$ was not varied in order to fix the origin in this noncentrosymmetric structure. Data used for this refinement were collected from crystal UW Museum no. 1769/3, using Fe$K\alpha$ radiation to avoid overlap of diffraction spots. A total of 512 monoclinic-merged and reindexed reflections was used. Several cycles of least-squares refinement were run, varying the scale factor and positions of atoms not fixed by symmetry. Refinement was stopped when bond lengths became unrealistic. The unweighted R at this point was 12%. Although the refinement could not be completed because of the presumed effects of twinning, the fairly low R factor of 12% (vs. 23% for the best six-layer structure) confirms the fact that the $3M_2$ stacking arrangement is basically correct for this lepidolite.

Correction for twinning

In order to correct the data for twinning, data were collected in the previously described manner from three different crystals using Cu$K\alpha$ radiation. It was assumed that the three crystals represented twinned crystals with only two individuals each and with different relative vol-

TABLE 3. Ideal atomic coordinates for the $3M_2$ structure

Atom type	x/a	y/b	z/c
M(1)	0.2500	0.7500	0.0000
M(2)	0.7500	0.9167	0.0000
M(3)	0.2500	0.0833	0.0000
O(1)	0.4287	0.9167	0.0360
O(2)	0.9287	0.0833	0.0360
OH(1)	0.9287	0.7500	0.0360
T(1)	0.4467	0.9167	0.0900
T(2)	0.9467	0.0833	0.0900
O(3)	0.4540	0.7500	0.1120
O(4)	0.7040	0.0000	0.1120
O(5)	0.2040	0.0000	0.1120
K(1)	0.9722	0.7500	0.1667
O(6)	0.1824	0.9167	0.2973
O(7)	0.6824	0.0833	0.2973
O(8)	0.6824	0.7500	0.2973
T(3)	0.4878	0.9167	0.2433
T(4)	0.9978	0.0833	0.2433
O(9)	0.4904	0.7500	0.2213
O(10)	0.7404	0.0000	0.2213
OH(2)	0.2404	0.0000	0.2213
M(4)	0.8611	0.9167	0.3333
M(5)	0.3611	0.7500	0.3333
M(6)	0.3611	0.0833	0.3333
O(11)	0.7064	0.7500	0.3693
O(12)	0.2064	0.9167	0.3693
OH(3)	0.7064	0.0833	0.3693
T(5)	0.7244	0.7500	0.4233
T(6)	0.2244	0.9167	0.4233
O(13)	0.4818	0.8333	0.4453
O(14)	0.9818	0.8333	0.4453
O(15)	0.2318	0.0833	0.4453
K(2)	0.7500	0.0833	0.5000

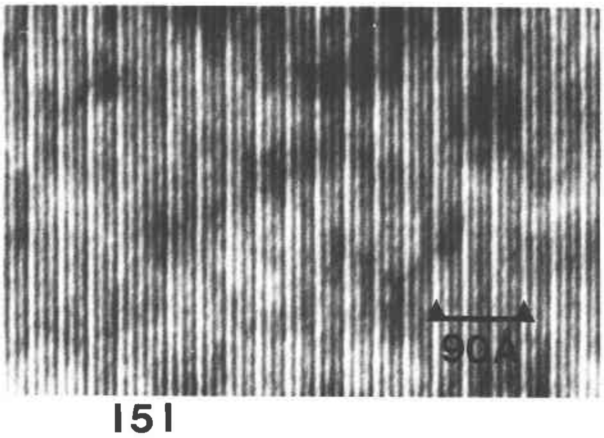

Fig. 4. Bright-field HRTEM image of three-layer polytype in Tørdal lepidolite. The three-layer periodicity is interrupted by a five-layer stacking defect, labeled "5."

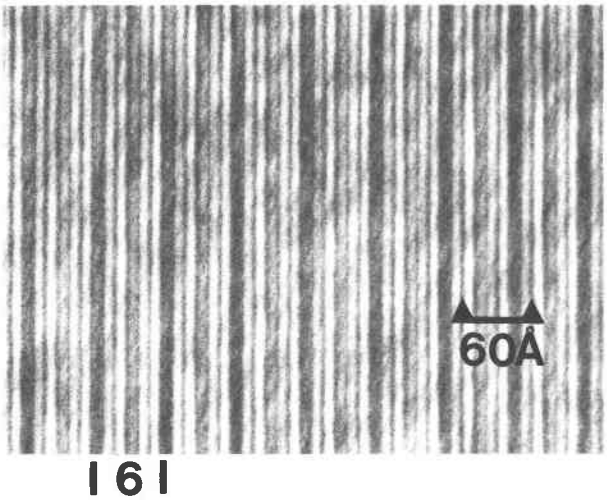

Fig. 5. Dark-field HRTEM image of six-layer polytype in Tørdal lepidolite, formed from the 02*l* diffraction row. One unit cell of the six-layer structure is labeled.

umes of the same type of twinning. The following steps were taken to achieve a corrected data set:

1. The volumes of the three crystals were put on the same scale by adjusting a scaling factor so that the intensities of each of the 00*l* reflections for all crystals were the same. All twin volumes should contribute to the 00*l* reflections. The intensities of all other reflections from each data set were then multiplied by the scaling factor appropriate for each crystal.
2. The relative proportions of twinning among the three crystals were determined by comparing the intensities of the "extra" six-layer–type reflections. Only one twin part contributes to these reflections.
3. A corrected "untwinned" data set was then produced by extrapolation to 0% twinning from the three data sets.

The corrected data set obtained in this way was used in a refinement of the $3M_2$ structure. Unfortunately, this "corrected" data set refined more poorly than uncorrected data sets. The most probable reason this method of correcting the data failed is that the twinning is more complex than originally assumed. More than one type of twinning or an intergrowth of three- and six-layer structures might be present.

TRUE NATURE OF THE TØRDAL LEPIDOLITE AS REVEALED BY TEM STUDY

Evidence related to twinning and stacking sequences was sought from TEM study. Specimens for the TEM study were prepared by ion milling of a 30-μm thin-section slice of the lepidolite cut normal to (001). The specimens were lightly coated with C. Electron microscopy was performed with a Philips 420 microscope operated at 120 kV, as described by Livi and Veblen (1987). High-resolution imaging was complicated by rapid beam damage of the mica and by warping of the thin edges of the specimen in the electron beam.

Selected-area electron-diffraction (SAED) patterns from some areas of Tørdal lepidolite show a three-layer periodicity (Fig. 3a), and some other areas indicate a periodic six-layer structure (Fig. 3b). In some other areas, an apparent six-layer periodicity in the diffraction pattern is interpreted as being due to twinning, because the diffraction spots are alternatively circular and elongate in shape, similar to the situation described previously for some of the crystals examined with X-ray diffraction. In addition, there are unequal spacings between spots along certain *hkl* rows (Fig. 3c). The twinning can be explained as stacking faults within a parent $3M_2 = [1\bar{2}1]$ crystal such that twin units are created by out-of-sequence rotations of ±60° or ±120° about the Z^* direction (equivalent to 180° rotation about an axis within the (001) plane for a centrosymmetric crystal). Some SAED patterns also show considerable streaking along the rows parallel to Z^*, indicative of disordered stacking with no true repeat along Z^* (Fig. 3d).

Previous studies involving the HRTEM imaging of micas have shown that under ideal conditions it is possible to determine the exact stacking sequence, at least in projection (Iijima and Buseck, 1978; Amouric et al., 1981). However, the Tørdal lepidolite of the present study exhibited very rapid beam damage and consistently warped in the electron beam, so that attempts to obtain two-dimensional structure images necessary for exact stacking-sequence determination failed. However, it was possible to obtain bright- and dark-field HRTEM lattice images by imaging the 00*l* and 02*l* reciprocal lattice rows, respectively; although they do not yield the exact stacking sequence, such images can be used to determine the periodicity of the structure in the stacking direction.

HRTEM images from some specimen areas show a good three-layer periodicity with occasional stacking faults (Fig. 4). In these areas, it was not possible to obtain six-layer periodicity in the images, even over a wide range of imaging conditions (crystal orientation, thickness, and microscope defocus), so that this can be assumed to be the $3M_2$ structure indicated by the X-ray study. The stacking faults produce individual two-layer, four-layer, five-layer, and wider lamellae that occasionally interrupt the sequences of true three-layer periodicity. In some areas, the intergrowth of lamellae of various widths is highly irregular, indicative of the severe stacking disorder suggested by some SAED patterns.

In other areas of the lepidolite, good sequences of six-layer periodicity can be obtained in HRTEM images (Fig. 5), although the appearance of the six-layer periodicity depends strongly on the exact imaging conditions used; the six-layer periodicity in the images grades directly along layers into pairs of apparent three-layer repeats. This three-layer subperiodicity probably appears because the *hkl*, $l = 2n$ diffraction spots are more intense than those with $l = 2n + 1$. The six-layer sequence observed in HRTEM images is assumed to correlate with the $6M_1$ structure indicated by X-ray study.

The TEM study thus shows that both three-layer and six-layer stacking sequences are present in the same "crystal," that stacking fault twins of the three-layer structure exist, and that material with substantial stacking disorder also is present. The TEM images do not reliably indicate the relative proportions of these various structures in the bulk sample, because of the relatively small amount of material used for SAED and HRTEM experiments.

The TEM results thus complement the X-ray diffraction results and explain why the structure of the Tørdal lepidolite could not be refined successfully on the basis of either a three- or six-layer model. The *R* values from the X-ray refinements suggest that the bulk of the Tørdal lepidolite is the $3M_2$ polytype. Because the "extra" six-layer diffraction spots are all very weak, none of these appear in the X-ray powder-diffraction pattern published by Bailey and Christie (1978).

EXPLANATION OF SECOND-HARMONIC ANALYSIS

As mentioned in the Introduction, a second-harmonic analysis of the Tørdal lepidolite by Horsey (1981) did not produce a signal. As in other tests for centrosymmetry, observation of a strong second-harmonic signal is proof of noncentrosymmetry, whereas a weak or zero signal does not necessarily prove a centrosymmetric structure. The negative result was of sufficient concern to generate the present restudy of the Tørdal lepidolite, however, as the $3M_2$ polytype is noncentrosymmetric. The Tørdal lepidolite is shown here to be an interleaving of a noncentrosymmetric $3M_2$ structure, a centrosymmetric $6M_1$ structure, twinned $3M_2$ regions that may simulate centrosymmetry, plus regions of disordered stacking. It is likely that the second-harmonic analysis did not detect the noncentrosymmetric $3M_2$ phase because of dilution by the centrosymmetric constituents.

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