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Direct Observation of Stacking Faults in Wollastonite

By

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Abstract

Wollastonite is well known to have diffuse streaks on *k* odd level in its X-ray diffraction photographs which have been said to be due to stacking fault with displacement of $b/2$ in stacking along the *a* direction (Jeffery, 1953). The stacking fault is directly observed by high-resolution electron microscopy. In the electron micrograph frequent stacking faults are observed in some places (faulting density $\alpha=0.4\sim0.6$), but the density varies very much from portion to portion. A dislocation is observed in two dimensional lattice image. Electron diffraction of wollastonite indicates some features which were not found in X-ray photographs; faint streaks along a^* on *k* even levels of (*hk*0) reciprocal lattice plane and faint streaks along c^* on *k* odd levels of (0*kl*) reciprocal lattice plane and fine structure of the streaks along a^* on *k* odd levels of (*hk*0) reciprocal lattice plane.

Introduction

Wollastonite is a silicate mineral classified structurally as pyroxinoid. Peacock (1935) concluded by optical and goniometric study that dimorph of calcium metasilicate exists: triclinic wollastonite and monoclinic parawollastonite. Ito (1950) suggested that both the triclinic and monoclinic varieties of calcium metasilicate could be constructed from hypothetical monoclinic cells (Space group $P2_1/m$) by shifting the succesive cells along the *a* direction in increments of $\pm b/4$. Further detailed work on wollastonite was done by Dornberger-Schiff *et al.* (1955) and Mamedov and Belov (1956), who indicated that it has infinite-chain of different type from pyroxene chain. Buerger and Prewitt (1961) have shown that the crystal structure proposed by Mamedov and Belov (1956) for wollastonite (Space group $P\bar{1}$) is correct and they attempted to explain some of the puzzling features of pyroxinoids. The crystal structure of parawollastonite was confirmed by Trojer (1968) as having space group $P2_1/a$. In addition to the polymorphs i.e. triclinic wollastonite (1T) and monoclinic parawollastonite (2M) which is a superstructure of 1T wollastonite (Jeffery, 1953; Dornberger-Schiff *et al.*, 1955), another superstructure of wollastonite (4T) was described by Wenk (1969). Wenk (1969) discussed the genesis of wollastonite polymorphs and emphasized the effects of stress and strain as well as of temperature and pressure. On the other hand Jeffery

(1953) first reported unusual X-ray diffraction phenomenon on wollastonite: continuous streaks in the a^* direction on k odd layer Weisenberg photographs. This phenomenon was interpreted by JEFFERY as due to 'mistakes' of $b/2$ in stacking from one lattice to another (completely stacking-disordered type). This corresponds to the 'hypothetical case' analyzed in detail by WILSON (1949). UEDA and TOMITA (1968) examined thirteen specimens from various localities in Japan and distinguished wollastonite polymorphs: wollastonite, parawollastonite and "Devon type modification" (completely stacking-disordered type).

Wollastonite often occurs with fibrous habit. It is difficult to make advanced research of finer crystalline textures of these specimens by X-ray single crystal or optical method, but electron microscopy and electron diffraction methods have become the potential methods to study these specimens. With these methods the characteristic features of fibrous wollastonite may be clarified in contrast with non-fibrous wollastonite.

Recently, electron microscopy containing high-resolution electron microscopy which gives directly the projected structure of the specimen as Fourier inverse transform of the electron diffraction pattern has become efficient in the study of fine textures of minerals (YADA; 1971, YOSHIDA and SUITO; 1972, BUSECK and IIJIMA; 1974). In the present study the author attempted to make direct observation of stacking fault with displacement of $b/2$ in stacking on (100) planes by high-resolution electron microscopy and to make detailed reaserch of fine scale crystalline texture of wollastonite by electron diffraction and electron microscopy.

Specimens examined and previous X-ray investigations

i) Specimen from Baba

The specimen examined in this high-resolution electron microscopic study occurs in contact zone at Baba, Kanta-cho, Fukuoka Pref., Japan and is the same one that was studied by X-ray single crystal method (UEDA and TOMITA, 1968). It is white in color, silky in luster and fibrous in shape. UEDA and TOMITA (1968) classified the specimen as "Devon type modification" (completely stacking-disordered type) with two sets of weak accessory spots on the streaks along the a^* direction on k odd levels: one set of the spots is placed on the positions corresponding to those of monoclinic form and the other set is located at middle points of the former. This diffraction pattern was interpreted as due to the stacking mistakes mentioned above and was explained in other words as a type of irregularity due to the presence of regions of wollastonite and parawollastonite.

ii) Other specimens

The specimens are mostly those examined by UEDA and TOMITA (1968), which were collected from the following localities: Aotani, Kashihara-shi, Osaka Pref.; Nakatatsu Mine, Izumi-mura, Fukui Pref.; Yoshihara Mine, Kitakyushu-shi, Fukuoka Pref.; Sannodake, Kawara-cho, Fukuoka Pref.; Yamato Mine, Mine-shi, Yamaguchi Pref.; Kotsubozawa, Rikuzentakata-shi, Iwate Pref.; Tsukide, Nishiasai-mura, Shiga Pref.; Gobesho, Shiga Pref. (Nyoigadake); Kamioka-cho, Gifu Pref.; Saimyoji, Himo-cho, Shiga Pref.; Kasuga-mura, Gifu Pref.

Experimental

Electron microscopes used were JEM-100C (mainly for high-resolution experiments) and JEM-7A (for electron diffraction and low magnification experiments). They were operated at 100 kV. Only a small piece (<1 mm) of wollastonite crystal aggregate was selected for the experiments, crushed, suspended in water and dispersed on perforated supporting films. Wollastonite fragments having proper thickness and orientation were selected in electron microscopy. Furthermore, for high-resolution electron microscopy critical beam alignment and stigmating of lenses were necessary and objective aperture used was 60 μ in diameter which corresponds to a radius of 2.5 $Å^{-1}$ in electron diffraction pattern. Trough-focus images were recorded at various focussing conditions and a little underfocus image gave good results.

Results and Discussions

I) Electron diffraction

i) Specimen from Baba

Even such fragments as picked up from a small piece (<1 mm) of wollastonite crystal aggregate gave somewhat varied (grain by grain) selected area ($\fallingdotseq 1\ \mu$) electron diffraction pattern of ($hk0$) reciprocal lattice plane (Fig. 1-a, -b, -c, -d.). Reciprocal lattice planes of wollastonite and parawollastonite are explained in Fig. 2. In Fig. 1-b continuous streaks along a^* on k odd levels were inferred to correspond to probability of faulting $\alpha \fallingdotseq 0.5$ by an optical diffraction experiment (Willis, 1958), and dense point row constructed from many weak diffraction maxima is dimly recognized as a fine structure of the streaks on k odd levels, which was not clearly observable in X-ray photographs presumably because of averaging effect. This may be due to such mode of statistically distributed mistakes as was found in devitrified glass and tobermorite (Bollmann, 1968). Furthermore faint streaks on k even levels are recognized, which has not been reported from X-ray experiments. This may be considered to be due to the disturbance in coherency along the a direction; for example, the displacement of stacking fault may not be precisely $b/2$ for some faults, or some disturbance concerning the fibrous character

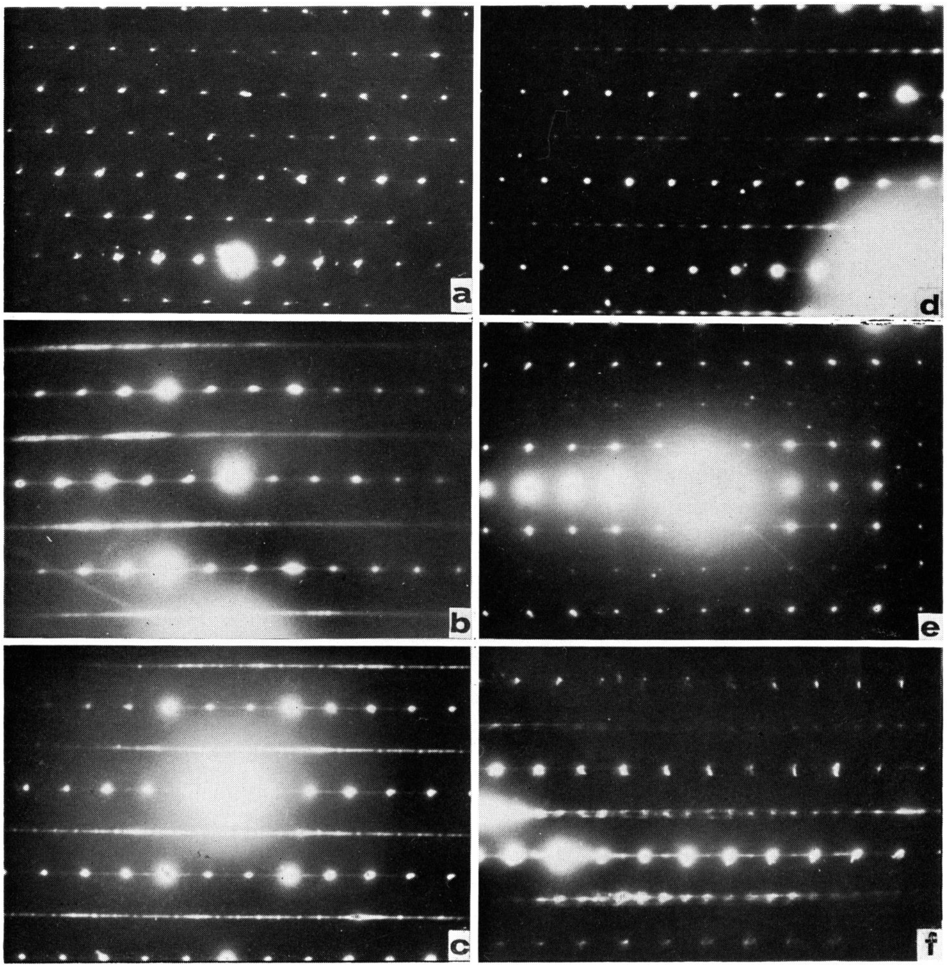

Fig. 1. Electron diffraction patterns of wollastonite (a): $(hk0)^*$, ordered triclinic wollastonite (1T), but with faint streaks on k odd levels. (b): $(hk0)^*$, completely stacking-disordered type, which consists of alternative rows of diffraction spots and streaks along a^* direction. As fine structure of the streaks on k odd levels, faint and dense diffraction maxima are recognized. Faint streaks between the diffraction spots on k even levels are observable (see text). (c): $(hk0)^*$ stacking-disordered type, with weak diffraction spots of right- and left-handed triclinic wollastonites (twinned) and parawollastonite. (d): $(hk0)^*$, twinned wollastonite. (e): $(0kl)^*$, faint streaks along the c^* direction are observed on k odd levels (see text). (f): $(hk0)^*$, parawollastonite from Kasuga Mine. Streaks along a^* direction.

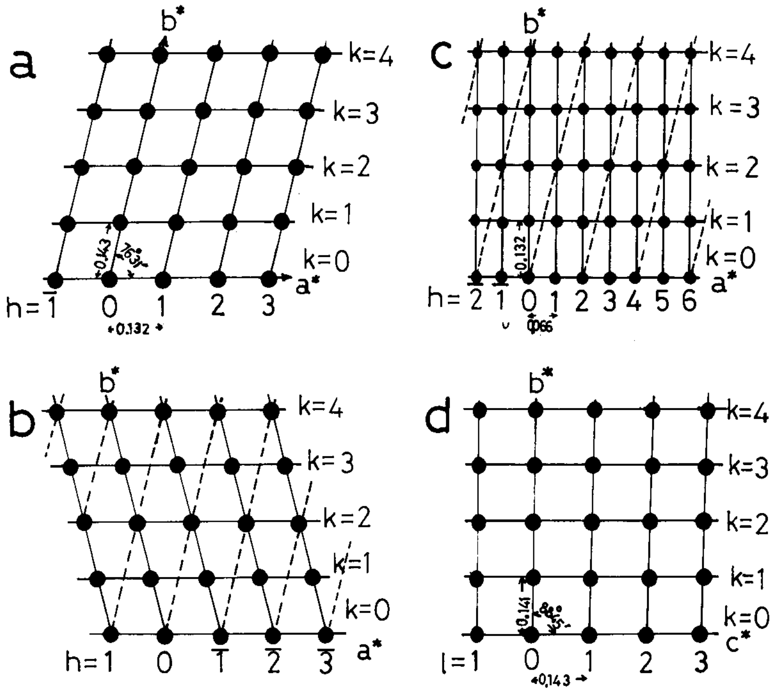

Fig. 2. (a), (b): Reciprocal lattice with right- and left-handed lattice orientation and reflections which should appear in electron diffraction of (*hk*0) reciprocal lattice plane of wollastonite. Twinned diffraction pattern is superimposition of (a) and (b). (c): Reciprocal lattice and reflections which should appear in electron diffraction of (*hk*0) reciprocal lattice plane of parawollastonite. Dashed lines in (b) and (c) represent the lattice net in (a). The composite of (a), (b) and (c) is found in Fig. 1–c. (d): Reciprocal lattice and reflections which should appear in electron diffraction of (0*kl*) reciprocal lattice plane of wollastonite.

of the specimen may be present. Fig. 1-c is almost the same diffraction pattern as X-ray photographs by JEFFERY (1953) and UEDA and TOMITA (1968). Faint streaks along c^* on k odd levels are sometimes observed in electron diffraction pattern of (0*kl*) reciprocal lattice plane (Fig. 1-e). This may indicate the stacking faults with displacement of $b/2$ in stacking from one lattice to another on (001) planes. The diffraction intensity of the streaks is usually very weak, so the region with stacking faults may be very small in the fragment. The whole single crystal also may have narrow regions with the stacking faults on (001) plane. Thus, wollastonite from Baba is considered to have frequent stacking faults on (100) plane and less frequent stacking faults on (001) plane. Besides electron diffraction

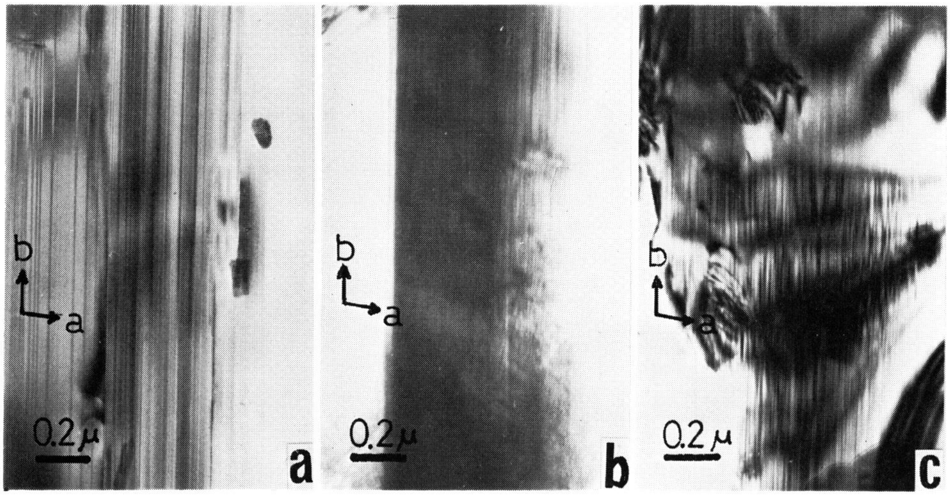

Fig. 3. Low magnification electron micrographs. (a): Wollastonite from Yoshihara Mine, whose electron diffraction pattern is triclinic normal type but with faint streaks along a^* direction. (b): Wollastonite from Baba, whose electron diffraction pattern is Fig. 1–b. (c): Parawollastonite from Kasuga Mine, whose electron diffraction pattern is Fig. 1–f.

patterns shown in Fig. 1 many other intermediate diffraction patterns of $(hk0)$ reciplocal lattice plane could be obtained. But in the specimen from Baba completely stacking-disordered type was relatively often obtained among the abovementioned varieties of electron diffraction patterns.

ii) Other specimens

Many varieties of electron diffraction patterns were usually taken even from the fragments of a small portion (<1 mm) of one specimen. The varieties of electron diffraction patterns observed up to the present are summarized in Table 1.

As a reference, pectorite specimen was examined by electron diffraction method. The specimen is from Sugashima, Mie Pref. and is of fibrous habit. The electron diffraction pattern of $(hk0)$ reciprocal lattice plane showed that pectorite also has normal type, twinned type and stacking-disordered type. Low magnification electron micrographs for some wollastonite specimens are shown in Fig. 3.

II) High-resolution electron microscopy on the specimen from Baba

High-resolution electron microscopy that is possible when thin crystal is suitably oriented so that the incident beam is parallel to a crystallographic axis, permits the imaging of the arrangement of unit cells and some of the details within the individual unit cells. In this study mainly two dimensional lattice images which could be directly correlated with structural details of wollastonite were taken.

Table 1 The varieties of electron diffraction pattern of wollastonites

Specimen Locality	Electron diffraction patterns of (*hk*0) reciprocal lattice plane			The streaks on *k* odd levels of electron diffraction pattern of (0*kl*) reciprocal lattice plane
	Triclinic (Wollastonite)	Monoclinic (Parawollastonite)	completely stacking-disordered type	
Baba	+	(+)	+	+
Aotani	+	+ normal parawo. and diso. parawo. (*k* odd lev.: with f. str.)		
Nakatatsu	+		+	
Yoshihara Mine	+ (*k* odd lev.: with f.∼w. str.)	+ (*k* even lev.: with v.f. str.)		
Sannodake		+ diso. parawo. (*k* even lev.: with w. str.)	+ (*k* even lev.: with v.f. str., *k* odd lev.: with wo. spots on str.)	+
Yamato Mine	+	+ (diffraction intensity of spots on *k* odd lev.: w.)	+	(+)
Kotsubozawa			+ (*k* odd lev.: with diffuse wo. spots, *k* even lev.: with v.f. str.)	
Tsukide	+	+ diso. parawo. (*k* odd lev. with diffuse spots, *k* even lev.: with v.f. str.)	+ (*k* odd lev.: with wo. spots, *k* even lev.: with v.f. str.)	+
Gobesho	+ normal wo. and twinned wo. (*k* even and odd lev.: with w. str.)		+	
Kamioka	+	+ diso. parawo.		
Saimyoji	+		+ (*k* even lev.: with f. str.)	+
Kasuga Mine	(+)	+	+	

+: present, lev.: levels, f.: faint, w.: weak, str.: streaks, wo.: wollastonite, parawo.: parawollastonite, diso.: stacking-disordered, v.: very.

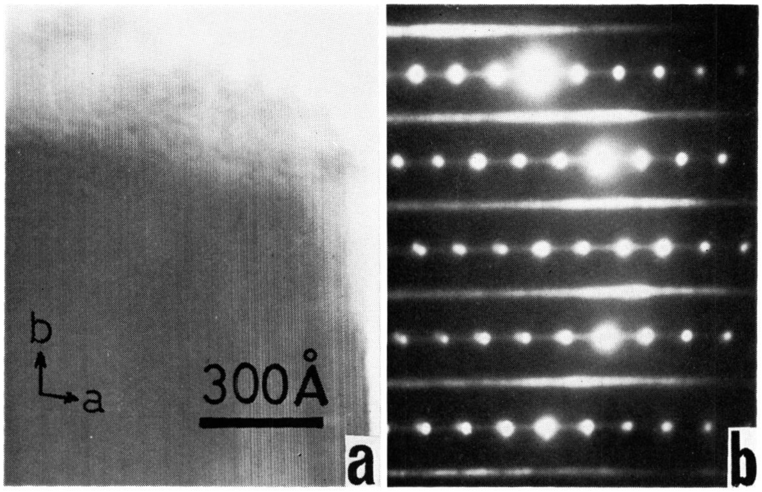

Fig. 4. (a): Edge of thin wollastonite fragment from Baba cleaved on {001}. (b): Electron diffraction pattern of the fragment (cf. Fig. 1–b)

Electron diffraction pattern (Fig. 4-b) of the fragment (Fig. 4-a) which originates from a fairly wide area of the specimen fragment exhibits completely stacking-disordered type in (*hk*0) reciprocal lattice plane (cf Fig. 1-b). As a whole the orientation of the fragment is approximately suitable. Fig. 5-a shows two dimensional lattice image in small portion near the edge of the fragment. The experiment of optical transform using He-Ne laser which performs the Fourier transform of high-resolution electron micrograph reduced to appropriate size is useful for checking the local orientation of the specimen. The optical transform (Fig. 5-b) of the photograph (Fig. 5-a) showed approximately symmetrical diffraction pattern; that is, locally the specimen is properly oriented (incident electron beam is parallel to *c*-axis). Unit cell scale is outlined in Fig. 5-a. The unit cells do not continue so long in the *a* direction and are frequently displaced by $b/2$ from one lattice to another. Hence, by direct observation, frequent stacking faults with displacement of $b/2$ in stacking on (100) planes were visibly ascertained. Abrupt contrast changes in the electron micrograph may be due to very local changes in crystal orientation and/or thickness. But in the range of the same contrast the information concerning the arrangement of unit cells may correspond directly to the real structures. The faulting density in this portion (Fig. 5-a) is 0.4. Electron micrograph of another portion is shown in Fig. 6-a (faulting density is 0.6). In the same fragment the faulting density varies strikingly from portion to portion. In some portion in the same fragment the unit cell arrangement in *ab* plane is almost perfect for a fairly wide area (>1000 Å along the *a* direction) (Fig. 7-a).

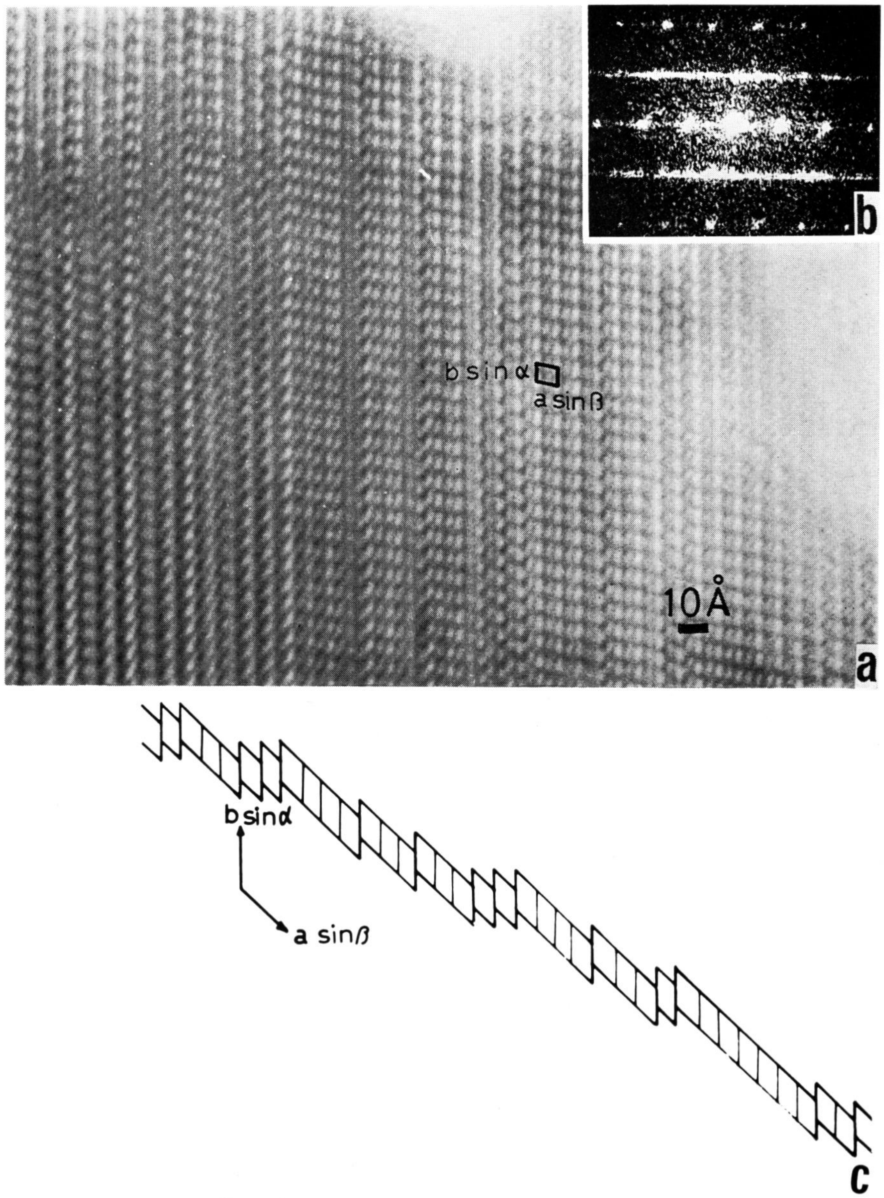

Fig. 5. (a): Two dimensional lattice image in small portion near the edge of the wollastonite fragment from Baba. Unit cell scale is outlined in the photograph. Frequent stacking faults on (100) plane are clearly seen. The faulting density is 0.4 in the area of this photograph. (b): Optical transform of the electron micrograph. The diffraction pattern is approximately symmetrical (incident beam is parallel to *c*-axis). (c) Schematical explanation of unit cell arrangement in the area of the photograph.

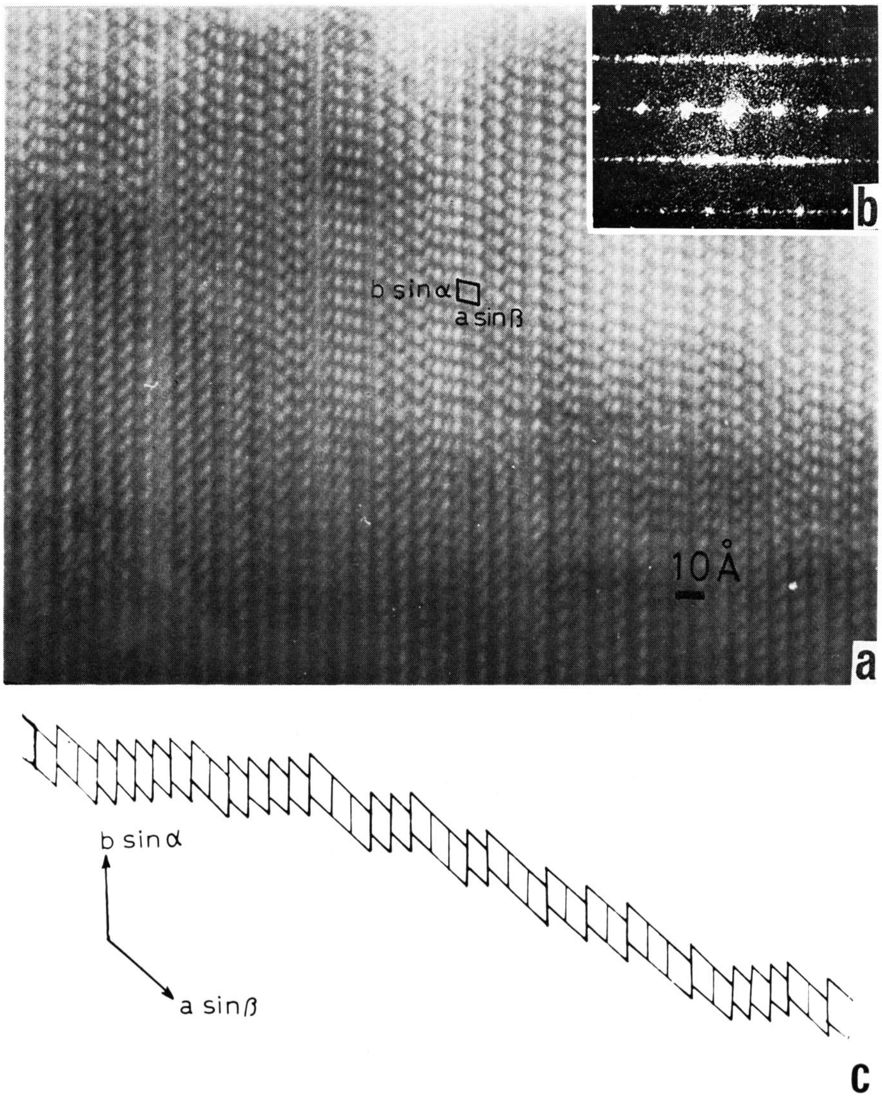

Fig. 6. (a): Two dimensional lattice image in small portion near the edge of the wollastonite fragment from Baba. More frequent stacking faults on (100) plane than in Fig. 5-a are seen. Faulting density is 0.6 in this portion. (b): Optical transform of the electron micrograph. The diffraction pattern is approximately symmetrical. (c): Schematical explanation of unit cell arrangement.

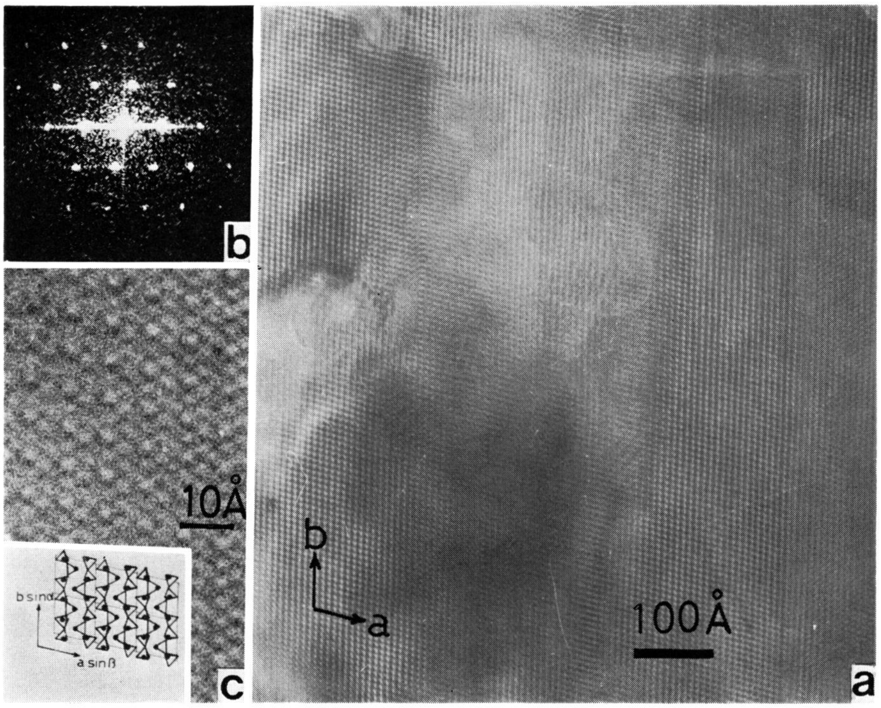

Fig. 7. (a): Two dimensional lattice image of wollastonite from Baba for a fairly wide area (*ab* plane). Almost perfect unit cell arrangement. (b): Optical transform of the electron micrograph. Symmetrical and normal diffraction pattern. (c): High-resolution electron micrograph of such portion.

High-resolution electron micrograph in such portion is shown in Fig. 7-c. Two point rows cross the unit cell parallel to *b*-axis and they may correspond to two wollastonite single chains in the unit cell.

The dislocation is shown by an arrow in Fig. 8.

Conclusion

Even small portion of wollastonite specimens from various localities gave various types of electron diffraction pattern of (*hk*0) reciprocal lattice plane. (Table 1) Faint streaks along the a^* direction on *k* even levels of electron diffraction pattern of wollastonite were observed, which may be due to the disturbance of coherency along the *a* direction. A fine structure of streaks of *k* odd levels of electron diffraction pattern of wollastonite was sometimes recognized as dense point row constructed from many weak diffraction maxima. Faint streaks along the c^* direction on *k* odd levels were observed on the electron diffraction pattern

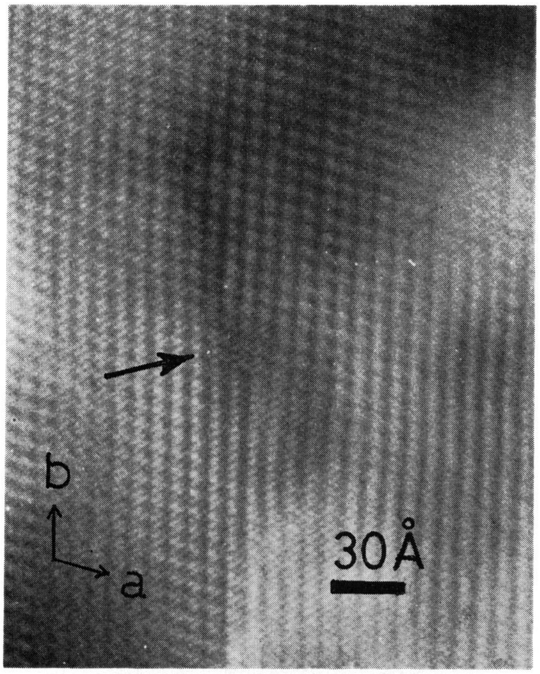

Fig. 8. Two dimensional lattice image of wollastonite from Baba (*ab* plane). The dislocation is indicated by an arrow. There exists a misfit between upper two lines and under one line along the *b* direction in the lattice image.

of (0*kl*) reciprocal lattice plane. This suggests stacking faults with displacement of *b*/2 in packing on (001) planes.

In two dimensional lattice image of wollastonite from Baba frequent stacking faults with displacement of *b*/2 in stacking on (100) planes were directly ascertained. The crystalline textures of the wollastonite were locally different even within one small fragment. A dislocation which is represented by a misfit between one line and two lines running parallel to *b*-axis in two dimensional lattice image is present.

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