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Microstructures of 18R martensite induced by deformation and thermomechanical cycles in CuZnAl shape memory alloy

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Microstructures of martensite in a Cu–26.4Zn–4.8Al shape memory alloy after deformation and thermomechanical cycles are reported. By detailed transmission electron microscopy investigation, it is clearly established that the microstructure induced by deformation is different from that induced by thermomechanical cycles, and the effects on the thermoelastic martensite transformation are also not the same at all. The microstructure of martensite induced by deformation is mainly deformation twins when the deformation exceeds the range of recoverable strain of the shape memory alloy. The main microstructure characteristic of martensite which has undergone thermomechanical cycles is regularly distributed dislocations, which are mainly aligned but sometimes tangled. Micro-twins similar to those found in deformed martensite and crossed bands also exist inside the martensite of samples after thermomechanical cycles, but the amounts are too small to be regarded as important microstructure features.

1. Introduction

The applications of shape memory alloys have been developing into many fields in recent years [1, 2]. In general the martensite first undergoes a large deformation when the shape memory effect (SME) is induced. It has been well established by optical microscope observation [3, 4] that the recoverable strain of shape memory alloys (SMAs) is achieved by the reorientation of martensite variants and is wholly different from that of plastic deformation, which is the result of dislocation slipping and twinning. However, few detailed investigations on the microstructure characteristics of martensite which has undergone the deformation process by reorientation have been made, and the microstructure of martensite when the deformation exceeds the reorientation stage also needs further clarification.

On the other hand, the applications of SME are in many cases associated with the two-way shape memory effect (TWSME) [1, 2]. The two-way behaviour is related to the formation of preferentially orientated martensite plates resulting from prior thermomechanical cycling of the shape memory alloys [5]. Till now, at least three different training procedures for developing a reversible two-way shape memory have been identified, i.e. the SME method [6], the SIM method [7], a combination of SME and SIM, and the method of heating the shape memory samples under constraint at a moderate temperature [8]. It has been shown that the method of combining SIM and SME procedures is the most effective [9]. The mechanism of TWSME obtained by the last method mentioned above has been clarified, and the α' plate formed during heating under constraint is considered to be responsible for TWSME [10–12]. However, concerning the mechanism of TWSME induced by thermomechanical cycles, i.e. SME, SIM or a combination of these, the explanation is still not very clear. In general it is considered that some residual stress exists in the form of lattice defects, which leads to the formation of preferentially orientated martensite variants [6, 7].

Concerning the lattice defects of two-way trained SMAs, most authors have focused their attention on the parent phase. It has been reported that there are dislocation tangles and "vestigial" martensite markings in the parent phase after thermomechanical training [13]. Generally, the Burgers vectors of dislocations in the parent phase are considered to be of $\langle 1\,1\,0 \rangle$ type [14]. Recently, a "comb-shaped" arrangement of dislocation was observed by Rios-Jara and Guenin [15, 16] in the parent phase of two-way trained CuZnAl [SMA], and the Burgers vector was determined to be $\langle 0\,0\,1 \rangle$ [15, 16] which is different from the generally accepted $\langle 1\,1\,0 \rangle$ Burgers vector [14]; however, the aligned dislocations are all proved to be along the $\langle 1\,1\,1 \rangle$ direction of the parent phase. It is certain that the formation of preferential variants is due to the existence of these dislocations, mainly in the parent phase. However, how the dislocations exist in martensite after the parent phase transforms to martensite is still unclear. In recent work, the authors of the present paper have reported a kind of dislocation microstructure in the martensite of a two-way trained CuZnAl alloy [17]. In the present investigation, we have studied the microstructures of martensite of a two-way trained alloy in more detail and hope to understand the behaviour of lattice defects in thermoelastic martensitic transformations more fully.

2. Experimental procedure

The alloy studied had a nominal composition of Cu–26.4Zn–4.8Al (wt %) and was melted under atmospheric pressure. The samples were step-quenched and the heat-treatment conditions and preparation of thin foils were as described elsewhere [18]. The M_s temperature was about 313 K. Samples of thin plate about 0.15 mm in thickness were further treated in two ways after heat treatment. One was solely deformation of martensite by tensile stress to different strains. The second was TWSME training by the combination of SIM and SME, i.e. thermomechanical cycles. About 30 cycles were taken and a stable TWSME was obtained. Any further heating and deformation was carefully avoided during thin foil preparation after the above two treatments in order to minimize the effects of other factors. The transmission electron microscope used was a JEM-100CXII operated at 100 kV with a $\pm 45°$ and $\pm 60°$ double-tilting specimen stage.

3. Results and discussion

The crystal structure and substructure of martensite and several kinds of crystal defect in the present alloy have been studied in previous work [18, 19]. The crystal structure of martensite is M18R and the substructure is mainly random stacking faults. The microstructure of martensite is greatly changed after the samples have undergone the deformation and thermomechanical cycles mentioned above.

3.1 Deformation microstructures of martensite

3.1.1. Deformation within reorientation stage

A series of specimens were chosen from the thin plate of fully martensitic phase stressed under tension to different strains. It is found that the reorientation process takes place when the strain is small. Comparing with observations by optical microscopy, however, some new features have been observed. Fig. 1a shows the morphology of martensite obtained after reorientation of martensite variants. It can be clearly seen that there are still marks of prior interfaces among unreorientated variants in some reorientated regions, as indicated by the arrows in Fig. 1a. These are called "marks of interface" in this work and they cannot be observed by optical microscopy. The "marks of interface" may be small-angle boundaries in nature, because the strong diffraction contrast of the two parts divided by them is reversed if a few degrees' change of the orientation of specimens is made. It has been reported that a martensite plate can grow across the small-angle boundaries [20] and the stacking faults are continuous through it. This implies that the martensite obtained after reorientation is not simply a single variant, but has small-angle boundaries inside it. Since the martensite can grow across the small-angle boundaries, it is expected that their existence in reorientated variants also does no harm to the SME. Another feature is that the reorientation of variants is often incomplete, as shown in Fig. 1b. It has often been found that there are some small residual variants, which may be too small to be observed under optical microscopy, in the reorientated region. There is a quite severe strain in the region neighbouring them and many dislocations have been formed. This also shows that the reorientation is not homogeneous in the whole sample.

3.1.2. Deformation exceeding the reorientation stage

When further deformation is applied to the reorientated martensite, new microstructure is induced. Fig. 2a shows the morphology obtained. It can be seen that many parallel narrow bands have been formed inside the large single variant resulting from the reorientation of several small variants. Selected-area electron diffraction analysis proved that these narrow

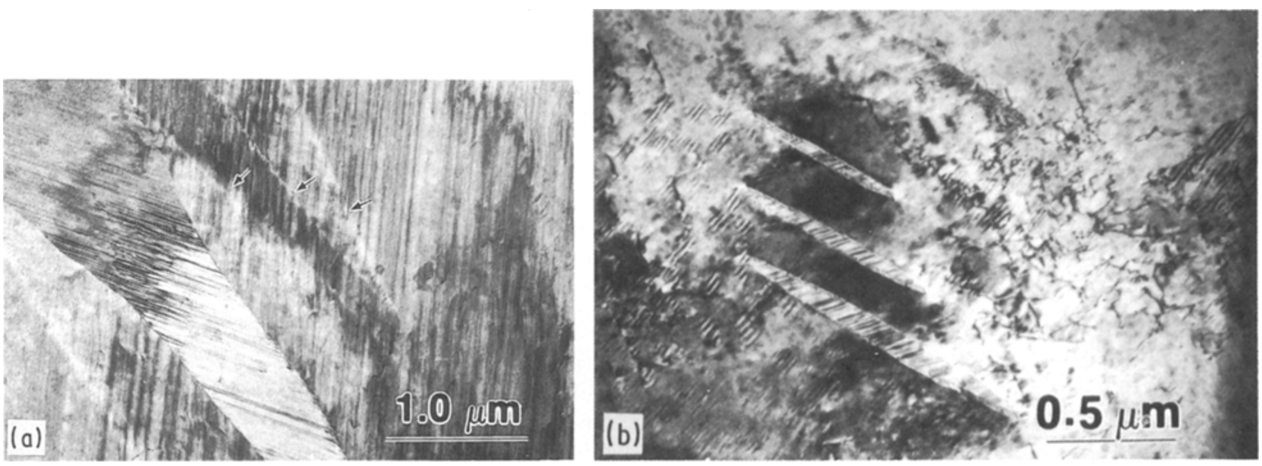

Figure 1 (a) Morphology of reorientated variants of martensite with "marks of interface". (b) Small residual variants existing in the reorientated region, with many dislocations formed near them in the matrix.

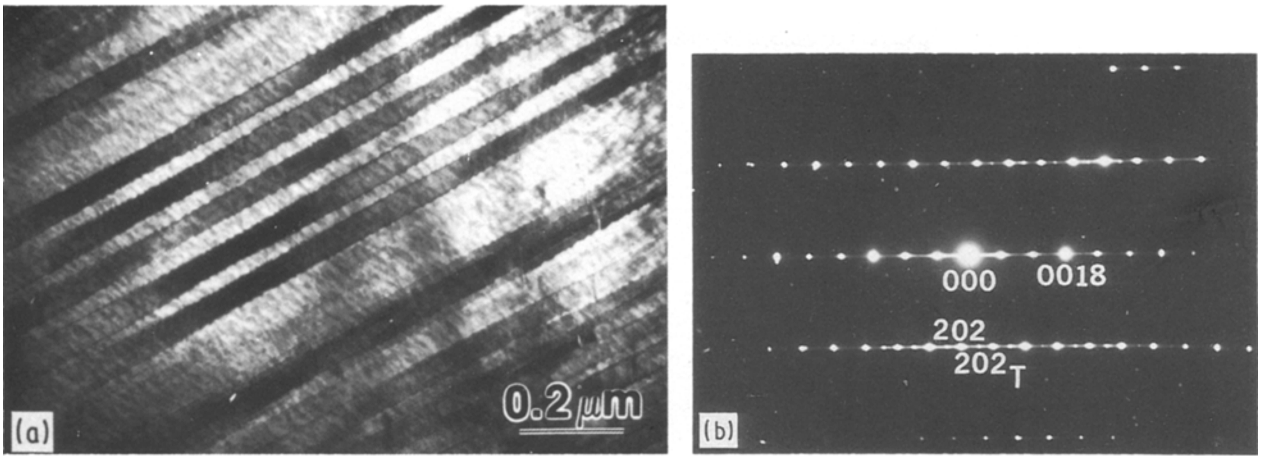

Figure 2 (a) Parallel narrow bands obtained by the deformation of martensite exceeding the reorientation stage. (b) Electron diffraction pattern corresponding to (a), showing the twin relations.

bands are in twin relationship with the martensite matrix, and the twin plane is (0 0 1) as shown in Fig. 2b. As the strain increases, the formation of deformation twins extends to the whole sample and its density also increases. Since the structure of martensite is a long-period close-packed structure with many random stacking faults, the stacking faults can be regarded as micro-twins of several atom layers. Adjoining atom planes may easily slip to the position of twins under the stress, and a micro-twin of several atom layers grows to a deformation twin of even more than hundreds of atom layers. This case is similar to that discussed in previous work [19]. It should be noted that dislocation microstructure has also been observed; however, the main microstructure of the deformed martensite is the deformation twins. Experiments have shown that the strain cannot be recovered to zero by heating to a temperature above A_f after the deformation twins have been formed. This means that the deformation contributed by the twinning is a plastic deformation.

3.2 Microstructure induced by thermomechanical cycles

3.2.1. Micro-twins inside the martensite matrix

It has also been observed that there is microstructure which is the same as that observed in the deformed martensite mentioned above, i.e. deformation twins. However, these deformation twins only exist in a few regions of the specimen; the whole amount and the width and density of it are also much smaller than that of the deformed martensite mentioned above. These micro-twins are also considered to be the result of plastic deformation of martensite. In the training process of TWSME (thermomechanical cycles), however, the deformation does not reach such a high level and the appearance of deformation twins may be the result of inhomogeneity of strain in the polycrystalline alloy in thermomechanical cycles, i.e. the twins are formed in the heavily deformed regions. Since deformation microstructure has been speculated to stabilize the defective martensite structure [21], the martensite plates containing many deformation twins would remain in the parent phase after reverse transformation as stablized martensite. As reported by Rapacioli *et al.* [22], stabilized martensite remaining in the parent phase results in a two-way shape memory effect. Although the stabilized martensite in their work was induced in a different way, martensite containing many deformation twins is expected to show a similar effect.

3.2.2. Crossed micro-bands inside the martensite plate

Besides the deformation twins mentioned above, another anomalous fine structure has also been found inside the martensite plate in a thermomechanical cycle trained sample, which is called "crossed micro-bands" in this work. Fig. 3 shows its morphology. It can be seen that some straight lines pass through the interface from one martensite variant to another. Fig. 3 was obtained when the incident beam direction was nearly parallel to the [0 1 0] direction of the martensite lattice. Selected-area electron diffraction analysis proved that the connected variants have an A–D relation, i.e. the two variants are in twin relationship with the (1 0 1 0) twin plane. By trace analysis it was determined that the striations along the direction indicated by a single arrow are parallel to the (0 0 1) basal plane of martensite, and these straight lines are generally regarded as stacking faults on the basal plane of martensite. However, the straight lines along the direction indicated by a double arrow pass across the interface from one martensite plate to another and form crossed micro-bands inside one plate. No such kind of substructure has been found in martensite. It was proved by trace analysis that the crossed micro-bands are parallel to the basal plane of the neighbouring variants. In fact, these micro-bands are continuous from the striations on the basal plane of the neighbouring variants.

The formation of crossed micro-bands is considered to be the outcome of two-way shape memory training.

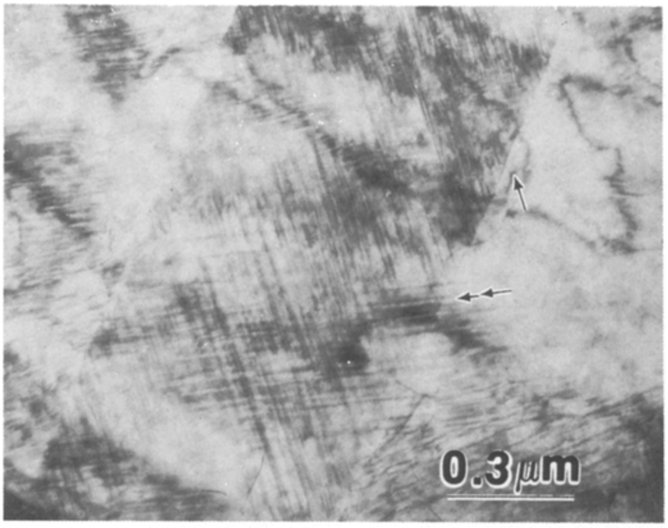

Figure 3 The morphology of crossed micro-bands inside a martensite plate (see text for details).

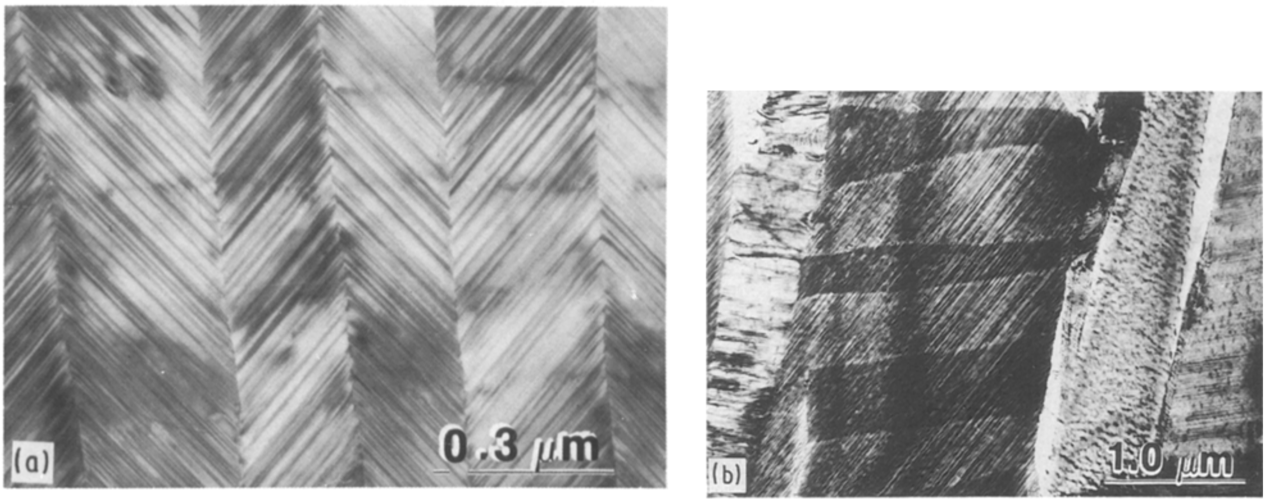

Figure 4 (a) Parallel martensite plates twin-related with the (1 0 1 0) plane. (b) A single variant obtained by the reorientation of parallel martensite variant pairs shown in (a).

As is well known, when samples are deformed in the martensite state, reorientation of variants takes place and a large single variant of martensite is obtained. Just as mentioned above, there are still "marks of interface" inside this large single variant. Fig. 4a shows the parallel martensite plates which are in (1 0 1 0) twin relationship, obtained in a sample without deformation and thermomechanical cycles, and Fig. 4b shows the situation after reorientation of the twin-related martensite plates shown in Fig. 4a. It can be seen from Fig. 4b that basal plane stacking faults have formed continuously through the "marks of interface". When the deformation exceeds the reorientation stage, it is expected that micro-bands may be produced by shear on the basal plane inside the variant obtained by reorientation, i.e. very fine deformation twins. When the martensite transforms back to the parent phase, the micro-bands may remain in the parent phase. When martensite forms in the following SIM and SME training cycle, the A–D variant pairs appear again and the micro-bands which had been on the basal plane of the single variant obtained by reorientation should be on two different planes of the two differently orientated variants. One is still on the basal plane and the other is nearly parallel to (1 0 1). Meanwhile the basal plane stacking faults certainly exist in both variants, so that there are crossed micro-bands inside one variant of the A–D variant pairs.

3.2.3. *Dislocation microstructures inside martensite*

Analogous to micro-twins, crossed micro-bands do not generally occur and dislocations (aligned and tangled dislocations) are the main microstructures of two-way trained martensite. Fig. 5 show some general morphology of the dislocation microstructure. It can be seen in Fig. 5a that the dislocations are all lying along one direction; they are therefore called "aligned dislocations". In some regions, however, the density of dislocations is very high and the aligned dislocations become tangled. Fig. 5b corresponds to such a situation. It has been proved that the aligned dislocations lie on the basal plane of the martensite lattice, and there are two typical distributions as shown in Fig. 5a and Fig. 5c. As Fig. 5a shows, two types of variant

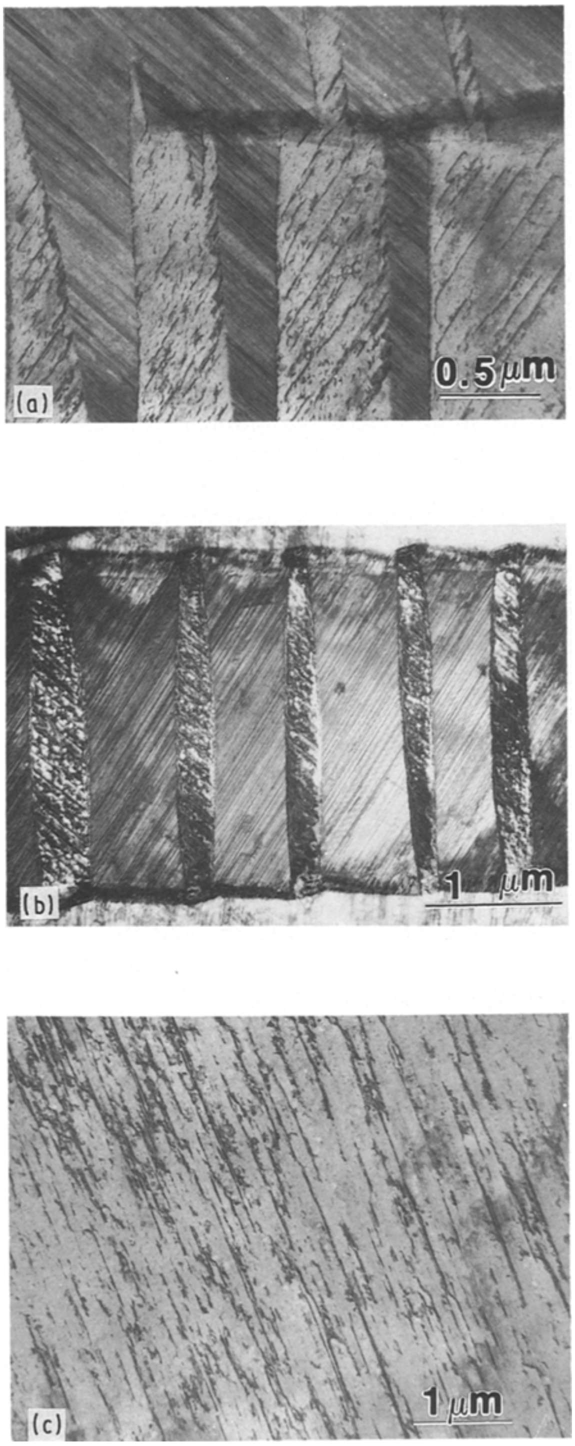

Figure 5 (a) Aligned dislocations in one type of variant of A–D variant pairs. (b) Tangled dislocations in one type of variant of A–D variant pairs. (c) Aligned dislocations in a large single variant.

which have the A–D relationship exist as parallel bands and the aligned dislocations exist only in one type of variant. Fig. 5c shows the second typical morphology of aligned dislocations. The aligned dislocations are formed in quite a large area which is a single variant, and there are no aligned dislocation microstructures in the variants adjacent to it. In both cases the stacking fault substructure has diminished after the dislocation microstructure formed inside martensite.

Since the aligned dislocations lie on the basal plane and they are all nearly straight lines, the direction of dislocation lines can be determined by obtaining the intersection of two planes containing aligned dislocations; or by tilting the foil to different orientations while keeping the (0 0 1) plane parallel to the incident beam, the incident beam direction is considered to be perpendicular to the dislocation line when the image of a dislocation is the longest. In the present work, the direction of aligned dislocations is determined as nearly along the [2 1 0] direction by the latter method, but its Burgers vector has not been determined yet and research on the dislocation structure in 18R martensite is still under way.

Fig. 6 shows another kind of microstructure induced by thermomechanical cycles. It is similar to that of aligned dislocations; however, the dislocations run through the whole martensite plate in the case of aligned dislocations, and in this situation the dislocations seem to exist inside the bands. The whole single plate of martensite is divided into several bands, and the dislocations exist in every other band. Another difference is that the dislocations are quite coarse and short in length in this case. They are much like the dislocation microstructures formed by cyclic deformation of copper single crystals, i.e. the dislocation ladder structure [23].

Since the aligned dislocations are widely distributed inside the martensite, they should be regarded as the main microstructure characteristic of two-way martensite and should be expected to play a key role in TWSME. As pointed out by Rios-Jara and Guenin [15] and Kajiwara and Kikuchi [14], many aligned dislocations have been found in the parent phase after a transformation cycle and thermomechanical cycles. It is expected that the aligned dislocations observed in this work correspond to the aligned dislocations in the parent phase observed by Rios-Jara and Guenin [15] and Kajiwara and Kikuchi [14], since the direction of dislocations in the parent phase has determined to be [1 1 1] [14, 15] and the [1 1 1] direction of the parent phase is parallel to the [2 1 0] direction of martensite [19]. This fact implies that the martensite containing aligned dislocation transforms back to the parent phase during heating, and the dislocations are inherited from the martensite by the parent phase. It is important that the existence of such dislocation substructure is not harmful to thermoelastic martensitic transformation. Since the dislocations have a regular distribution in the parent phase, which is dependent on the training process, the martensite variant should be formed in a regular orientation during cooling below the M_s temperature which is coincident with that formed in the SIM and SME training procedure; a macroscopic shape change induced in the training process by strain would be established automatically during cooling below M_s.

Concerning the results presented in this paper, the microstructure induced by thermomechanical cycles is quite complex. The formation of these crystal defects, their detailed structure and the relations among them are still unclear and need further study.

4. Conclusions

The microstructures of martensite induced by deformation and thermomechanical cycles in Cu–26.4Zn–4.8Al alloy have been studied by means of transmission electron microscopy. The conclusions of the present investigation can be summarized as follows:

1. The deformation microstructure of martensite is mainly deformation twins with (0 0 1) as the twin plane when the strain exceed the reorientation stage. The prior interface between martensite variants often remains as "marks of interface" inside the single variant obtained by reorientation, and the reorientation is often incomplete.
2. The microstructure induced by thermomechanical cycles includes several kinds of crystal defect; they are "micro-twins", "crossed micro-bands" and several types of dislocation. Micro-twins are similar to the deformation twins of deformed martensite, and micro-bands are crossed with basal plane stacking faults, nearly on the (1 0 1) plane. However, they are not often observed and should not be regarded as the main microstructure of two-way trained martensite.
3. The aligned dislocations distributed regularly inside martensite are a characteristic feature of the microstructure of two-way trained martensite. It has been determined that they are along the [2 1 0] direction of martensite and expected to correspond to aligned dislocations along the [1 1 1] direction in the parent phase. Sometimes they appeared as tangled dislocations inside variants.
4. All the above microstructures in martensite are speculated to remain in the parent phase after the martensite transforms back to parent phase, which would lead to the formation of certain preferential variants of martensite.

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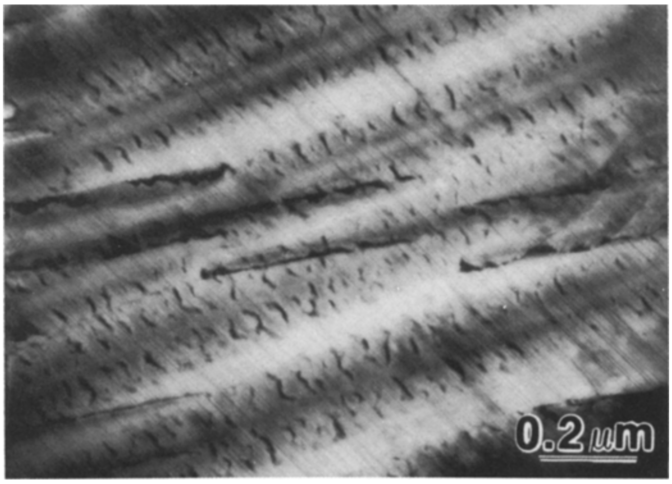

Figure 6 Dislocation walls inside a single variant of martensite (see text for details).

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