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The Application of the Kossel Technique and Electron Microscopy to the Study of the Microstructure of Fe-32% Ni Martensite Crystals

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The back-reflection Kossel technique and transmission electron microscopy have been used to investigate the internal structure of Fe-32% Ni martensite crystals. The internal structure of these martensites has been shown to consist in general of interpenetrating bands of fine twins and slip dislocations on the (112) and (101) planes respectively. A small number of plates have been shown to exhibit interpenetrating {112} and {145} twinning. The relevance of complex internal martensite structures to the crystallography of martensitic transformations and to the deformation of internally twinned martensitic phases in general are discussed.

1. Introduction

The product phase of a martensitic transformation often exhibits internal faulting consisting of slip dislocations, stacking faults or fine twinning which is usually assumed to be representative of the lattice-invariant deformation associated with the transformation, but which may also be a result of the accommodation distortion which a martensite plate experiences as it grows. In either case the nature of the microstructure will influence the observed habit plane and orientation relationship between the parent and product phases, as well as contributing to the strength of the martensite crystals [1] since it is clear that internal twinning on a fine scale, for example, must influence considerably the operative deformation modes in martensite.

Previous investigations of the microstructure of ferrous martensite crystals have included transmission electron microscopy studies of thin films. Unfortunately it is not always possible to relate the valuable high resolution studies of the microstructure obtained in this way to the observed crystallographic features of martensite plates formed in bulk material, and thus to assess the generality of the microstructure. Extensive optical microscopy studies of etch traces delineating the microstructure of martensite crystals have been of limited value because subsequent trace analysis cannot be readily related to the martensite crystal lattice, owing to the difficulty of determining the full orientation of martensite crystals having widths of only tens of microns. However, the application to this problem of the back reflection Kossel technique, which utilises a microprobe analyser [2, 3], makes it possible to determine the full orientation of martensite plates with widths as small as five microns, and accurately to relate this information to a two-surface analysis of etch-traces.

This technique has been used as the basis of the present investigation of the microstructure of martensite crystals in an Fe-32 wt % Ni alloy from which good quality Kossel patterns are obtained. Detailed electron microscopy studies of the microstructure of martensite crystals in this alloy have also been made and the results of both studies for each particular type of microstructure are presented simultaneously in section 3.

The experimental procedures used in this investigation are briefly described in section 2. The relevance of the type of microstructure observed to the deformation studies of martensite crystals and to the crystallographic aspects of martensitic transformations is discussed in section 4.

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2. Experimental Procedure

The experimental results presented in section 3 are restricted to those obtained from Fe-32% Ni alloys prepared from 4N pure materials by induction melting under a vacuum of 10^{-5} torr. Specimens cut from single crystal materials grown by the standard Bridgman technique were mechanically polished and etched electrolytically in 10:1 acetic-perchloric electrolyte. In this investigation the etch-trace analysis was supplemented by bright-field and dark-field transmission electron microscopy of thin slices, prepared by the standard Bollman technique, of as-transformed single crystal material.

Valuable information concerning the internal structure of martensite plates can be obtained from etch studies of martensite plates in bulk specimens. Such information is quantitative in nature only when the trace analysis, and in particular a full two-surface analysis, can be related directly to the martensite crystal lattice. The problem of determining the orientation of the small martensite crystals was readily overcome in the present investigation by using the back-reflection Kossel technique to determine the full orientation of the martensite plates. The divergent beam of X-rays required for the formation of the Kossel patterns were generated within the specimen using an AE1-SEM2 electron probe microanalyser [2, 3]. An example of a typical Kossel pattern obtained from an

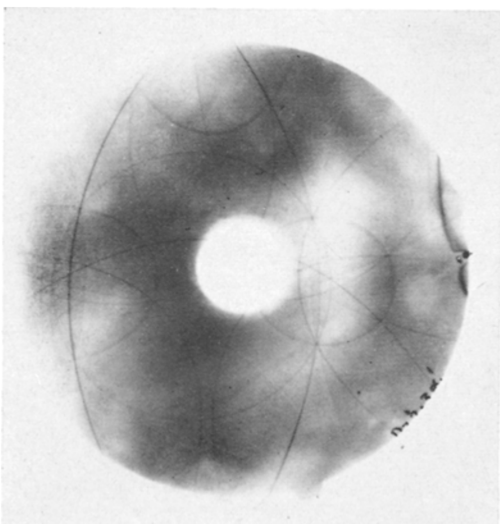


Figure 1 A typical back-reflection Kossel pattern obtained from an Fe–32% Ni martensite crystal reduced 40% for reproduction.

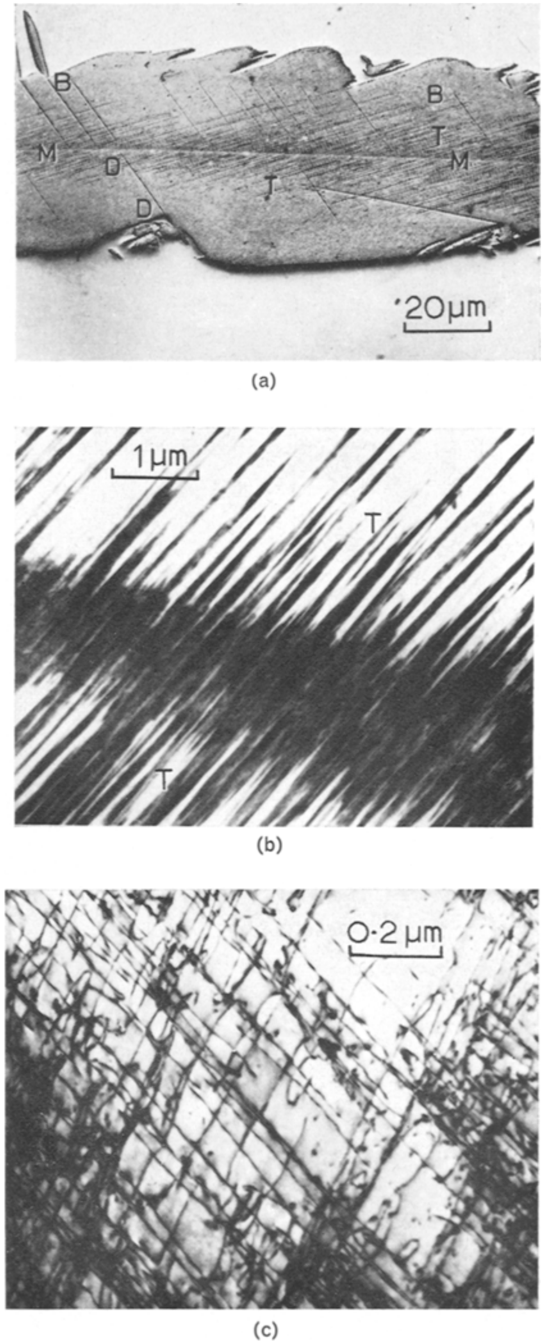


Figure 2 (*a*) Optical micrograph showing classical micro-etch structure of Fe–32% Ni martensites. Etch traces T–T delineate transformation twins. (*b*) Transmission electron micrograph of an Fe–32% Ni martensite plate showing a comparable microstructure of transformation twins to that indicated by fig. 2*a*. (*c*) Transmission electron micrograph illustrating a typical dislocation substructure which exists outside the twinned region in a martensite plate.

Fe-32% Ni martensite plate is shown in fig. 1. Rapid solution of such patterns is achieved by the use of previously calibrated charts [4].

3. Experimental Results

The optical micrograph of an etched martensite plate in fig. 2a illustrates the classical type of micro-etch structure of acicular martensite plates in ferrous alloys [5]. The etch-traces T-T delineate transformation twins and define a (1 1 2) plane with respect to the martensite lattice basis. Fig. 2b is an electron micrograph of a comparable microstructure. Both micrographs of the Fe-32% Ni alloy show that the transformation twins are most dense in the central (midrib) region of the martensite plate. Bands of slip dislocations can be observed in the regions of plates which are not internally twinned, as shown for example in fig. 2c. The micrographs 2a to c are comparable with micrographs published by many authors in the type of microstructure they portray. However, no definitive optical evidence as to the nature of the slip plane associated with slip dislocations observed outside the midrib region has been published. The most recent electron-microscopical determination of the slip plane [6] indicates that the slip plane is {1 1 2} which differs from the {1 1 0} plane postulated in a much earlier study [7].

Detailed studies of a large number of as-transformed martensite plates in an Fe-32% Ni alloy using the procedures described in section 2 have shown that many martensite plates exhibit a micro-etch structure delineated by two sets of interpenetrating traces, T_1-T_1 and T_2-T_2, as shown in fig. 3a. The martensite plate shown in fig. 3a is plate 1 of the four martensite plates shown in fig. 3b, each of which exhibit two sets of internal traces. Plates 1 and 3 are the same crystallographic variants, as are plates 2 and 4. All four plates are crystallographically equivalent however, as the relative orientations of etch-traces T_1-T_1 and T_2-T_2 in each case can be described by $(1 1 2)_b$ and $(0 1 1)_b$ respectively, if each martensite habit plane is indexed in the same way with respect to the martensite basis. Since twinning on $(0 1 1)_b$ in body-centred cubic materials is not possible, the second set of etch-traces, T_2-T_2, must be due to etch-pits delineating dislocations on the $(0 1 1)_b$ planes. This result has been confirmed by electron microscopy. The relative orientations of the transformation twins, T_1-T_1 $(1 1 2)_b$, and the slip bands, T_2-T_2 $(0 1 1)_b$, in the as-transformed martensite plate shown in fig. 4a correspond exactly with the results of the optical microscopy. Detailed analysis of the micrograph showed that the "cut-off" direction [5], indicated by the broken line in fig. 4a, is the $[1 1 \bar{1}]$ direction. This result is consistent with the segmentation of transformation twins by slip dislocations propagating on the (0 1 1) planes. Fig. 4b is a more typical example of this frequently observed structure, where again the "cut-off" direction is a $\langle 1 1 1\rangle$ direction.

In addition to martensite plates exhibiting two sets of etch-traces defining $(1 1 2)_b$ and $(0 1 1)_b$ planes, the authors have also observed martensite plates exhibiting two sets of etch-traces defining $\{1 1 2\}_b$ and $\{1 4 5\}_b$ planes. An optical micrograph illustrating this structure is

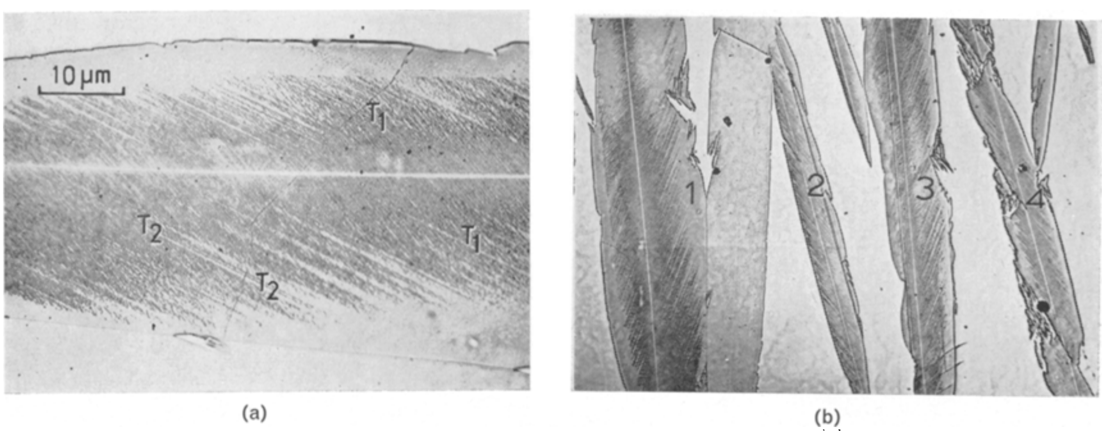


Figure 3 (a) Optical micrograph of an etched martensite plate exhibiting two sets of etch-traces. (b) Optical micrograph showing four martensite plates exhibiting two sets of internal etch-traces. All plates are crystallographically equivalent and a high magnification micrograph of plate I is shown in fig. 3a.

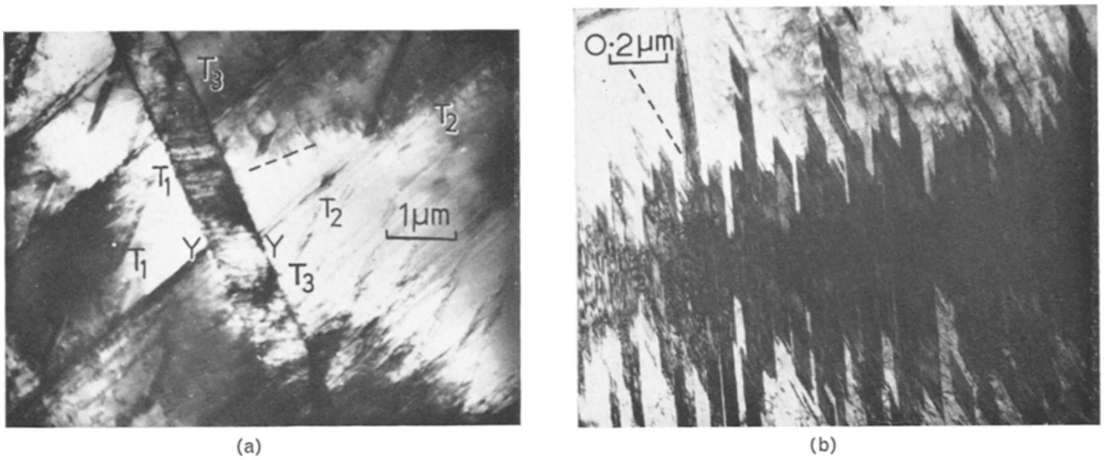


Figure 4 (a) Transmission electron micrograph showing a comparable structure to that represented by fig.3a. T_3–T_3 is parallel to a deformation twin, T_1–T_1 is parallel to transformation twins and T_2–T_2 is parallel to bands of slip dislocations (b) Transmission electron micrograph showing transformation twins exhibiting a "cut-off" direction parallel to the broken line.

shown in fig. 5a. The traces T_2-T_2 in fig. 5a are much coarser than the traces T_2-T_2 in fig. 3a, and the morphology of the former is consistent with them being twin traces. Additional confirmation that traces T_2-T_2 are twin traces is given by fig. 5b, which shows deformation twins parallel to T_2-T_2. The deviation of the deformation twin traces at the centre of the plate is attributed to the interaction of the deformation twins with the (1 1 2) transformation twins which are most dense at the centre of the plate. The relative variants of the twins T_1 and T_2 in fig. 5a, as determined by the procedure described in section 2, are (1 1 2) and (5 1 4) respectively.

A similar structure has been observed electron-microscopically, as shown in figs. 6a and b which are bright- and dark-field micrographs of a martensite structure exhibiting two sets of fine twins which extended over a large volume of the martensite plate. The trace T_3-T_3 in fig. 6 delineates a deformation twin. Two electron diffraction patterns were obtained from the field illustrated in fig. 6a, a [1 1 5] pattern from the matrix M, a [1 1 1] pattern from the twin T_2-T_2 and T_3-T_3. The diffraction patterns describe an orientation relationship between the twins T_2-T_2, T_3-T_3 and the matrix M as a rotation of π about the $[\bar{1}11]$ direction in the matrix crystal. In the stereographic projection analysis of fig. 6c of this situation the trace

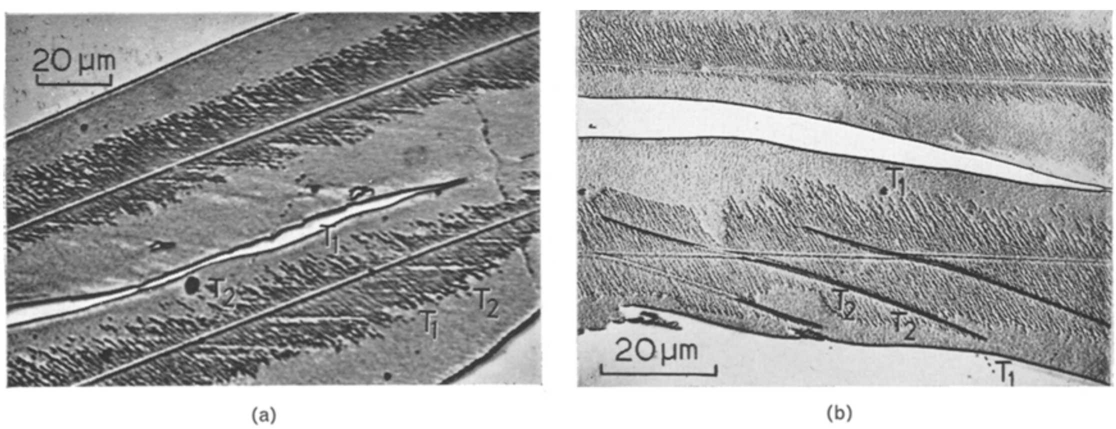
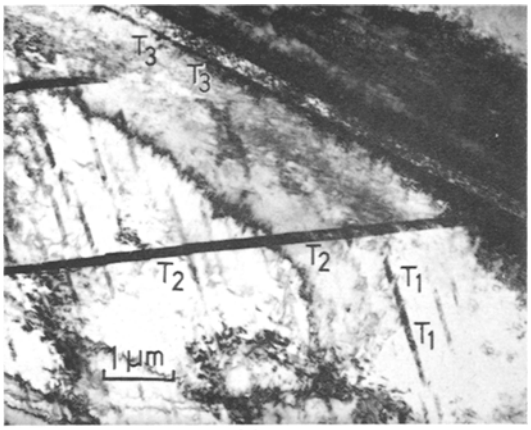
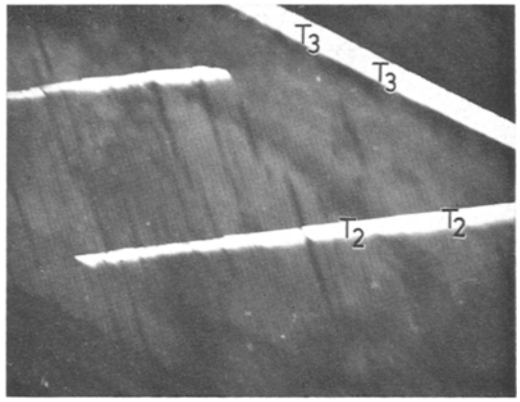


Figure 5 (a) Optical micrograph of an etched martensite plate exhibiting two sets of etch-traces defining two sets of interpenetrating twins. (b) Optical micrograph of a field adjacent to that shown in fig. 5a showing deformation twins parallel to traces T_2–T_2 in fig. 5a.



(a)



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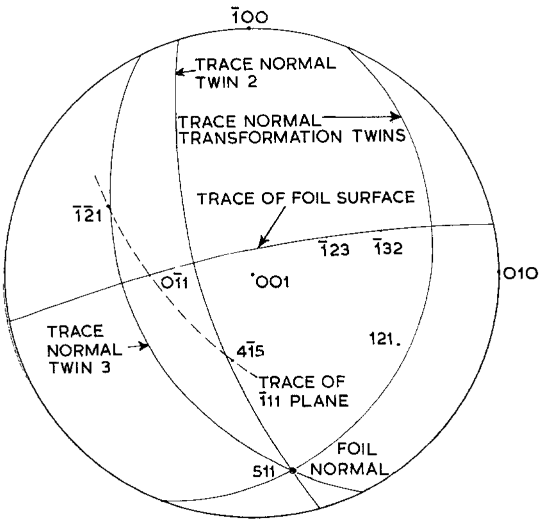

(c)

Figure 6 (*a*) and (*b*) Bright-field and dark-field electron micrographs respectively of an Fe–32% Ni martensite plate exhibiting two sets of interpenetrating fine twins T_1–T_1 and T_2T_2. T_3–T_3 is parallel to a deformation twin. (*c*) Stereographic projection analysis of figs. 6*a* and *b* which utilises the electron diffraction patterns obtained from the field illustrated in figs. 6*a* and *b*.

normals to the three sets of twins are plotted together with the traces of the ($\bar{1}$11) plane. The trace normal to twin T_3 intersects the ($\bar{1}$11) plane at a point within 1° of the ($\bar{1}\bar{2}$1) pole, indicating that the deformation twin has a ($\bar{1}\bar{2}$1) composition plane.* The trace normal to twin T_2-T_2 intersects ($\bar{1}$11) at a point within 2° of the (4$\bar{1}$5) pole indicating that these twins are not {112} twins but twins with a composition plane close to (4$\bar{1}$5). Relative projected widths of the twins confirmed this result, and also that the twins T_1-T_1 have a composition plane (1$\bar{2}$1) as indicated by single surface trace analysis. The relative variants of [145] and [112} twinning in the examples of multiple internal twinning presented in figs. 5 and 6 are different, but these examples show quite conclusively that twinning on a fine scale other than [112} twinning can occur in martensite plates in Fe-Ni acicular martensite plates, and that the composition plane of these twins is of the form [145].

4. Discussion

It is apparent from the results presented in section 3 that varied types of fine structure can be operative in Fe-32% Ni martensite crystals, and it is reasonable to assume that a similar diversity of fine structures be observed in most ferrous martensites. This is substantiated by the recent detailed electron microscopy study of the microstructure of the body-centred tetragonal Fe-0.82% C and Fe-1.82% C martensites by Oka and Wayman [9]. The scatter in habit planes of ferrous martensites has often been attributed to a multiple-shear lattice-invariant deformation, and recently two generalised theories of martensite crystallography have been published [10, 11] which can incorporate such a

*In general there are 24 different ways of describing an orientation relationship between two cubic crystals. In relating two electron diffraction patterns from possibly twin related crystals we choose the orientation relationship to be that of a rotation of π about a direction, and this direction following the classical laws of deformation twinning [8], is taken to be the shear direction associated with the twinning process. The plane normal to this direction must contain the normal to the shear plane associated with the twin and it is for this reason that the ($\bar{1}$11) plane has been introduced into the trace analysis.

deformation in a physically realisable way. The fine structures observed in the present study would at first examination tend to support the concept of the multiple-shear lattice invariant deformation. The scatter in martensite habit plane poles for martensite plates in a single austenite grain is shown in fig. 7. The martensite

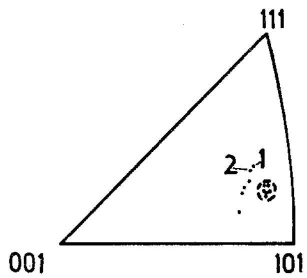

Figure 7 Stereographic projection showing the variation of habit planes of martensite plates in a single austenite grain with internal structure.

plates which exhibit a single set of well-defined etch-traces have poles which fall within the broken line in fig. 7. These plates are the first formed plates of the transformation and are associated with little scatter. All plates exhibiting two sets of well-defined etch-traces have poles which fall outside the broken line and give rise to the large scatter in martensite habit planes for martensite plates in a single austenite grain. These plates are the later formed plates. Some examples are shown in fig. 3b and the pole of plate 1 in fig. 3b, for example, is indicated by a "1" in fig. 7. The set of etch traces in addition to those delineating (112) transformation twins have been shown to delineate {101} planes. In some martensite plates these additional traces are well-defined, but it must be stressed that, when using the etching technique described in section 2, most plates are observed to exhibit to some degree two sets of fine traces. The dislocations associated with a lattice-invariant deformation would not be expected to be observed throughout the volume of a martensite plate [9]. One must therefore conclude from this study that the scatter in habit plane, which can be related to the presence of a well-defined second set of etch-traces delineating bands of slip dislocations, cannot be described by the results of the application of the phenomenological martensite crystallography theories in their present form.

We attribute the presence of bands of slip dislocations within a martensite plate to the accommodation distortion which a martensite plate experiences as it grows. This postulate is consistent with the uniform distribution and increased density of slip dislocations observed in the later formed plates and also with the formation of "deformation" twins in as-transformed Fe-32% Ni martensite plates at a well-defined stage in the growth of a particular plate. An example of this frequently observed form of twinning [12] is shown in fig. 2a, where the ends of a family of twins D-D delineate a "boundary" B-B which is parallel to the midrib trace M-M. The formation of a non-planar martensite-austenite interface may clearly be related to the occurrence of these twins.

Few martensite plates have been observed that exhibit {145} transformation twins, but the evidence that {145} twinning is operative is quite conclusive and is most likely to be described by the twinning mode [13].

$$K_1 = \{145\},\ K_2 = \{\bar{1}03\},\ \eta_1 = \langle\bar{1}\bar{1}1\rangle,\ \eta_2 = \langle 351\rangle\,.$$

The magnitude of the shear strain of this mode is 1.871, which is extremely large but may be compensated for by the facts that the Burgers vector of the twinning dislocations is $\frac{1}{6}\langle 111\rangle$, which is the same as for {112} twinning; the energies of the {145} twin boundary is approximately the same as that for the {112} boundary [14], and also that no shuffles are associated with the shear process. The habit plane pole of the martensite plate exhibiting {112} and {145} twinning shown in fig. 5 is indicated in fig. 7 by a "2", and is seen to differ widely from the other habit plane poles indicated. The low frequency of occurrence of this type of plate does not really permit a meaningful comparison of experimental results with the results of an application of the "multiple-shear lattice-invariant deformation" martensite theories [11, 12]. The application of these theories to this problem must in any case be open to question because the Bain correspondence matrix gives no {145} planes in the product which are derived from mirror planes in the parent [9].

The results of the investigation described in this paper show quite conclusively that the structure of Fe-32% Ni martensites consists in general of interpenetrating (112) transformation twins and (011) bands of slip dislocations. This fine structure must influence the operation of deformation modes in ferrous martensite crystals, as must the transformation structure influence the deformation of martensitic phases of many other crystalline materials [15]. The important question to be answered is whether or not the primary modes of deformation, which are slip and deformation twinning, can propagate through the fine structure. Very few detailed experimental investigations of the propagation of either mode of deformation through twinned structures have been carried out. The geometry of the incorporation of pre-existing slip dislocations by deformation twins in body-centred cubic metals, β-tin, mercury and the close-packed hexagonal metals have been considered by Sleeswyk and Verbraak [16], Ishii and Kiho [17], Guyoncourt and Crocker [18] and Yoo [19] respectively. Yoo [20] has also listed the experimental results which confirm that slip dislocations can decompose, on penetrating a twin boundary, into an integral number of unit twinning dislocations and a slip dislocation in the twin crystal. Direct confirmation by transmission electron microscopy of this process has been provided by Tomsett and Bevis [21, 22] for the interaction of basal slip dislocations with $\{10\bar{1}2\}$ twins in zinc. The study most relevant to the problem of the interactions of slip dislocations with a transformation structure has been given by Chilton and Kelly [1], the limited extent of this investigation being related to the difficulty of carrying out such a study in ferrous martensites. The most notable early work on the interaction of deformation twins with twins is due to Cahn in a study of deformation twinning in alpha-uranium [23], by Sleeswyk [24] and more recently by Levasseur [25] on deformation twinning in bcc metals. The authors of the present paper, in a detailed study of deformation twinning in Fe-32% Ni and Fe-23% Ni – 0.6% C martensites [12], have shown, however, that a size effect exists and that the mechanism by which deformation twins propagate through other deformation twins, which are in general coarse twins, is different from the propagation of deformation twins through fine transformation twins. In conclusion we will describe some of the general features of the propagation of deformation twins through fine twins in an Fe-32% Ni martensite which should have some application to other crystalline materials which are twinned on a fine scale. The general process of the deformation of martensite plates by deformation twinning is illustrated by fig. 4a which shows a deformation twin T_3-T_3 propagating through bands of slip dislocations and transformation twins.

Examination of many hundreds of twinned ferrous martensite plates [12] has shown that deformation twins propagate undeviated by the fine structure, which – as there is a non-uniform distribution of transformation twins – must mean that deformation twins propagate undeviated by the transformation twins. An example is given by the "deformation" twins D-D in fig. 2a whereas one of the two exceptions observed by the investigators is shown in fig. 5b. The mechanism of propagation of deformation twins must be either by detwinning of the transformation twins, a mechanism proposed by Sleeswyk [24] in terms of emissary dislocations and the incorporation of slip dislocations by deformation twins, for by secondary twinning [26], the mode of deformation being determined by the relative orientation of deformation twin and transformation twin. The deformation twin T_3-T_3 of fig. 4a propagates by detwinning of the transformation twins as shown by fig. 8, which is a micrograph of a region adjacent to that shown in fig. 4a and is of exactly the form expected by the Sleeswyk detwinning mechanism. The slip dislocations must also be incorporated by the deformation twin [14] and in fig. 4a are parallel to the line Y-Y within the deformation twin.

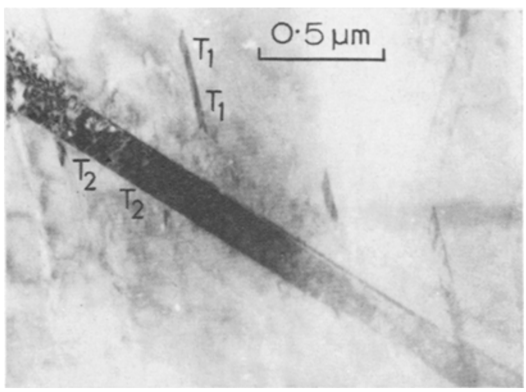

Figure 8 Transmission electron micrograph of a region of a martensite plate adjacent to that shown in fig. 4a illustrating a deformation twin-transformation twin interaction of the Sleeswyk detwinning type.

An example of secondary twinning is shown in the electron micrograph of fig. 9. The deformation twin T_2-T_2 has propagated, undeviated by the transformation twins T_1-T_1; the transformation twins on opposite sides of the deformation twin have undergone a relative displacement

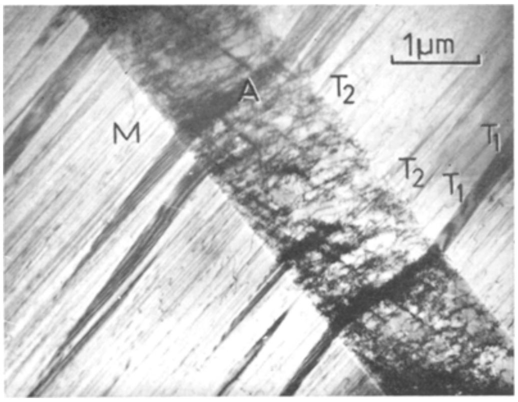

Figure 9 Transmission electron micrograph showing a deformation twin which has propagated by secondary twinning of the transformation twins.

determined by the magnitude of the twinning shear of the deformation twin, and the orientation of the regions of transformation twins incorporated by the deformation twin (regions A in fig. 9) differ in general from that of the original transformation twin, deformation twin and matrix martensite (M). It has been possible [12] to relate the observed deformation twinning modes in ferrous martensites to those which are able to propagate by simple mechanisms in one of the two modes described above.

It is clear that two ways of studying the type of problem discussed above are desirable. A study on a macroscopic scale is required to assess the frequency of occurrence and general morphological features of a particular deformation mode. This has been made possible by the direct application of the back-reflection Kossel technique. In addition, electron-microscopic studies are required for confirmation of the nature of the interaction of a particular mode with the fine structure.

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