

PROGRESS IN METAL PHYSICS 3

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BRUCE CHALMERS, D.Sc., Ph.D.

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PROGRESS SERIES

METAL PHYSICS

3

FOREWORD TO VOLUME I

THE study of the physical properties of metals has developed through a number of stages. The first was that in which the mechanical properties were correlated empirically with the heat treatment to which the metal had been subjected and, sometimes, to the chemical composition. At this stage the successful treatment of metals was an art, in the sense that experience rather than understanding led to the most satisfactory results. The next stage, in which the internal structure of the metal was examined, was based originally on the use of the microscope and it was found that many experimental facts could be explained in terms of effects that were of the right size to be seen under magnifications of less than about two thousand. The development of the x-ray diffraction techniques allowed phenomena of a smaller order of magnitude to be examined, and much of the existing information was found to be comprehensible in terms of the geometry of the crystal structure of the various phases that were visible under the microscope.

More recent development can perhaps best be discussed by a division of the field into what may be termed 'statics' and 'dynamics'. Under the former heading is the study of the conditions which govern the structure of a metal or alloy when it is in thermodynamic and mechanical equilibrium. The theories of the phases that are present in equilibrium and of the elastic constants have made remarkable progress in terms of rapidly developing theory of the part played by electrons on the metal.

Under the heading 'dynamic' effects we may include both the conditions governing the approach to equilibrium in respect of the phases that are present, in which diffusion plays an important part, and the response of a metal to forces which are sufficient to cause non-recoverable or plastic mechanical deformation.

These and associated subjects have advanced so rapidly that it has become difficult for research workers in one part of the field to remain up to date in other branches. It is the purpose of these volumes, which will appear as an annual series, to present authoritative reviews of the present state of knowledge in specialized aspects of the field that includes both physical metallurgy and metal physics. It is not intended that any one volume should form a complete textbook on these subjects. It is hoped rather that a few subjects of current interest should be discussed rather fully so as to cover, in the course of several years, all the more important aspects in which progress is being made. In order to make the series reasonably self contained it is proposed that the necessary 'historical' background should be included the first time a particular subject is discussed. Subsequent articles on such subjects will generally only cover the more recent progress.

B. CHALMERS

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1

CRYSTALLOGRAPHY OF TRANSFORMATIONS

J. S. Bowles and C. S. Barrett

A WEALTH of information has been accumulated during the last twenty years on the crystallographic aspects of phase transformations in the solid state. The purpose of this paper is to give a general review of this work and to consider in detail the contribution it has made to our knowledge of the atom movements accompanying the transformations, in the transforming material, in the boundaries of the transforming regions and in the surrounding matrix.

It is customary to classify the transformations in metals into two types. In the one type, the so called martensitic transformations, the new crystal is formed from the parent crystal by atom movements which are so cooperative and regular that they produce a change in the shape of the transforming region. There is little or no interchange in the position of neighbouring atoms during the transformation. In the second type, the nucleation and growth transformations, the new phase grows from a nucleus by thermally activated diffusion of atoms from the parent phase to the new phase. Interchange of position of neighbouring atoms does occur as they cross the interphase boundary and there is no change in shape of the transforming region.

This classification is somewhat arbitrary, for the characteristics that have been chosen as the definitive ones may not represent the most fundamental differences between the two types. The final selection of the most appropriate definitions and names for the two types of transformations awaits a more complete understanding of the transformation mechanisms. The choice of terms used here is justified by their widespread use in the past. Martensitic transformations have also been called diffusionless and athermal; these names emphasize the idea that the transformation involves no thermally activated transfer of atoms at the interphase boundary and no composition change in the transforming region. Such names, however, encounter the objection that diffusion within the parent phase itself may in some cases be a controlling factor even though diffusion at the boundary is not. Nucleation and growth transformations have also been termed diffusion processes and thermal transformations, names which bring out the occurrence of thermally activated transfer of atoms to the growing phase.

Much attention in the present discussion is directed to the atom movements and lattice distortions in the two transformations, but this

is not intended to imply that these are necessarily the most convenient criteria for distinguishing in the laboratory one type of transformation from the other. An exceedingly useful criterion for this purpose is the fact that nucleation and growth processes go to completion isothermally, whereas martensite transformations rapidly come to a halt when the rate of cooling or heating is brought to zero, and the transformation process is resumed only if the temperature change is resumed. Other characteristics are listed in the following section.

MARTENSITE TRANSFORMATIONS

General Characteristics

The distinctive features of martensitic transformations, which have been reviewed in detail by TROIANO and GRENINGER¹ and others,²⁻⁶ may be stated concisely.

- 1 Individual martensite crystals of plate shape, or of lens shape as in *Figure 1*, form on crystallographic planes. In most alloys the crystals form within times of the order of 10^{-4} seconds or less, even at low temperatures. The transformation occurs without change of composition of the transforming regions. Some examples have been observed in which the plates broaden relatively slowly, but even in these the original plate is observed to form very quickly (the indium-thallium transformation is an example⁷).
- 2 Transformation occurs only when the specimen is cooling and there is negligible transformation if cooling is stopped. In most instances the reaction proceeds only by the formation of new plates, not by continued growth of plates already formed.
- 3 The transformation begins at a temperature (M_s) that is not depressed by increasing the cooling velocity, but is dependent upon composition and prior thermal and mechanical history.
- 4 The low temperature phase reverts to the original high temperature phase in a similar manner during heating, unless competing reactions set in, such as nucleation and growth of the original phase.
- 5 The martensite crystals form in many specific orientations within a parent crystal, but on heating these revert to the original single crystal in which they formed.
- 6 Visible tilting and distortion of a polished surface is caused by the transformation.
- 7 Plastic deformation is effective in forming martensite above the M_s temperature, provided the temperature does not exceed a critical value (M_d). Cold work tends to suppress the transformation when a cold-worked specimen is subsequently cooled.

But cooling while a stress is applied increases the amount of transformation at each temperature, according to SCHEIL⁸ and to CHANG and READ.⁹ McREYNOLDS¹⁰ found no effect on M_s of stresses that were purely elastic.

- 8 Stabilization of the high temperature phase results in some alloys if cooling is sufficiently slow or if a specimen is held for a sufficient time in a suitable temperature range; the result of stabilization is to lessen the amount of transformation that occurs during cooling to a given temperature.
- 9 The crystal structure of a strain-induced martensite is not necessarily the same as that of the martensite which forms spontaneously in the same specimen, and the structure may vary slightly or even change completely with varying composition of the parent phase. Two different martensitic phases may co-exist in an alloy, as in copper-aluminium,¹¹ copper-tin¹² and iron-15 per cent manganese.¹³
- 10 Transformations to hexagonal close-packed or face-centred cubic structures and to some others have been found to yield an imperfect form of the structures in which faulty stacking of the atomic layers is frequent. This occurs in cobalt, where the transformation from face-centred cubic to hexagonal close-packed produces faults in the stacking of the basal planes on the average about ten to fourteen planes apart,¹⁴ and in lithium, where the body-centred cubic to hexagonal close-packed transformation produces even more closely spaced faults.¹⁵ Strain-induced precipitation in copper-silicon alloys is productive of much faulting and the same is true of certain alloys of the silver-tin and silver-antimony systems.¹⁷ Each of these systems provides conditions such that faulty stacking does not require the formation of interfaces of high energy.
- 11 The curve of amount transformed as a function of temperature on cooling below M_s is not retraced on heating, for rapid reversion during heating begins only at temperatures higher than M_s . Thus there is marked temperature hysteresis of a magnitude that depends upon the strain energies and interface energies involved. Cooling again after partial reversion brings a new M_s into play (M_s') which is somewhat higher than the original M_s .¹⁸

Diffusionless Characteristics—It is generally agreed that martensite transformations do not involve any interchange in the positions of pairs or rings of neighbouring atoms, the total displacement of any atom relative to its neighbours being smaller than the atomic radius. Of the considerable amount of experimental evidence available to substantiate

this view, possibly the most convincing is the persistence of ordered arrangements of atoms throughout the transformation. It is found, for example, that if the parent phase contains a superlattice the martensitic phase also contains a superlattice. The martensite transformation in the copper-aluminum alloys offers an example of this behaviour.¹⁹

This behaviour also applies to ordered arrangements of atoms in interstitial solid solution. It has been pointed out by LYMAN²⁰ and by JASWON and WHEELER²¹ that the arrangement of carbon atoms on the interstitial sites in the body-centred tetragonal martensite of iron-carbon alloys is precisely the arrangement which is expected if the carbon atoms do not move from the interstices they occupy in the parent phase, face-centred cubic austenite.

In austenite the carbon atoms are situated at the midpoints of the cube edges and in the equivalent positions that are at the centres of the cubes. However, the face-centred cubic structure referred to different axes is identical with a body-centred tetragonal structure having an axial ratio of $\sqrt{2}$; referred to these axes the carbon atoms in austenite occur at the midpoints of the four longer edges of the unit cell and at the centres of the square bases, which are equivalent positions. It has been shown^{22, 23} that these are also the positions occupied by the carbon atoms in body-centred tetragonal martensite so that the transformation from austenite to martensite can be regarded simply as a distortion of one body-centred tetragonal structure into another, the interstitial carbon atoms remaining in the same positions relative to the neighbouring iron atoms. This inheritance of the position of carbon atoms is undoubtedly the explanation of the tetragonality of martensite for in their inherited positions all carbon atoms tend to stretch every unit cell in a crystal of martensite in the same direction. If the carbon atoms had moved into all interstices at random during the transformation, the situation would be different—the resulting structure would be body-centred cubic.* The essential point is that not all equivalent interstices in a body-centred cubic structure are derived from equivalent interstices in the face-centred cubic structure.

Further evidence suggesting that martensite transformations only involve small displacements of atoms relative to their neighbours is found in the change in shape of the transformed volume. If a polished surface is prepared on a sample of the parent phase the transformation produces characteristic upheavals on the polished surface which effectively render the martensitic phase visible. These relief effects

* ZENER²⁴ has pointed out that martensite retains its tetragonality for times which are very much longer than the average time (about one second at room temperature) that a carbon atom stays in a particular interstitial position. He proposes that this is due to the martensite being an ordered interstitial superlattice, at a temperature below the critical ordering temperature *i.e.* the tetragonal unit cell has lower free energy than the cubic cell that would result from disordering.

will be considered in more detail later. For the present it will be sufficient to point out that any transformation in which the atoms do not interchange position must be accompanied by a change in shape of the transformed volume and it is this change in shape that is responsible for the observed relief effects.

The remaining evidence consists of observations which indicate that the activation energy for the unit process in the transformation, whatever its nature, is very small compared with the activation energy for diffusion at the same temperature. From this it is concluded that the atom displacements are smaller than an interatomic distance. These observations are 1 martensite transformations are not accompanied by any change in composition so there is no need for long range diffusion. 2 Martensite plates form with extreme rapidity even at temperatures at which the mean time of stay of atoms on the lattice points is very long. The times of formation of martensite plates in a 71 : 29 iron-nickel alloy and in zirconium have been measured by FÖRSTER and SCHEIL²⁵ who found values of less than 0.7×10^{-4} sec and 3.4×10^{-4} sec respectively. The maximum possible speed is of course the speed of propagation of a sound wave in the material and it may well be that this is the actual rate of formation. In any event, the transformations are in many cases accompanied by an audible click. This is true even at temperatures near 4°K.²⁶ 3 Martensite transformations do not proceed at constant temperatures as a thermally activated process does, but cease when cooling is arrested.*

Atom Movements—Since martensite transformations are accomplished without interchange in the positions of neighbouring atoms, it follows that the observed orientation relationships between the parent structure and the martensitic structure, and the observed change in shape of the transformed volume, are a direct consequence of the atom movements that occur during the transformation. The atom movements can therefore be deduced from the crystallographic data.

The first attempt to describe the shifting of atoms in a martensite transformation was made by BAIN,²⁷ who proposed a mechanism for the transformation from face-centred cubic austenite to body-centred tetragonal martensite in steel. Bain noted that this transformation could be regarded, as was pointed out previously, as a distortion of one body-centred tetragonal structure with an axial ratio of $\sqrt{2}$, into another having the axial ratio of martensite. He therefore proposed that the transformation merely involves a compression of the *c* axis, and an expansion of the *a* axis until the martensite axial ratio is reached. Bain also proposed from the observed dependence of the axial ratio on carbon content, that it is the interstitially dissolved carbon atoms which prevent the axial ratio from going completely to unity.

* However, see the comment on this point in the footnote on p 22.

Table I. Orientation Relationships and Habit Planes in Martensite Transformations

System	Composition	Transformation on Cooling	Orientation Relationships	Habit Plane in Parent Phase	Remarks and References
Fe-C ..	0.0-4% C	FCC \rightarrow BCT	Presumably as for 1.4% C	Laths \parallel $\{110\}\gamma$ arrayed on the $\{111\}\gamma$ planes	28, 29 This has been referred to as a $\{111\}$ habit plane but is probably just a further degeneration of the $\{225\}$ plates into needles
	0.5-1.4% C	FCC \rightarrow BCT	$\{111\}\gamma \parallel \{110\}\alpha$ $\langle 110 \rangle \gamma \parallel \langle 111 \rangle \alpha$ This is known as the Kurdjumov-Sachs relationship	With decreasing C% the plates degenerate into laths with axis $\parallel \langle 110 \rangle \gamma$ (259)	28 32 Orientations by pole figure method
	1.5-1.8% C	FCC \rightarrow BCT	Unknown		28 Lattice relationships have not been determined in this range. A deviation from the low carbon relationship is suspected from the difference in habit plane
Fe-Ni ..	27-34% Ni	FCC \rightarrow BCC	$\{111\}\gamma \parallel \{110\}\alpha$ $\langle 211 \rangle \gamma \parallel \langle 110 \rangle \alpha$ This is known as the Nishiyama relationship	Approx. (259) but wide scatter	28, 32-34 Orientation determination by pole figure method. The Kurdjumov-Sachs relationship is found above room temperature and Nishiyama below, according to 34
Fe-Ni-C ..	1.2% C 11.5% Ni 0.8% C 22% Ni	FCC \rightarrow BCT FCC \rightarrow BCT	As in Fig. 4	Approx. (259) Approx. (259)	28 35 Precision orientation determination
Fe-Mn ..	1-15% Mn 13-25% Mn	FCC \rightarrow BCC (α') FCC \rightarrow CP Hex. (ϵ)	Unknown Unknown	Unknown Unknown	13 13
Cu-Zn ..	40% Zn 40% Zn + 1% Pb + Sn	BCC \rightarrow Structure unknown BCC \rightarrow FCT	Unknown Unknown	(155) or (166) Unknown	36 37
Cu-Sn ..	25% Sn	BCC \rightarrow Structure unknown	Unknown	(193)	36

Cu-Al	..	11-13.1% Al	BCC $\rightarrow \beta'$ β' is a distorted CPH structure with the $[001]$ 2° from normal to the (001) plane. Angle between (100) and (010) differs by 1° from 120° BCC \rightarrow CPH γ' The CPH phase becomes distorted into an orthorhombic lattice with increasing Al%	As in Fig. 5	Approx. (133) 12° from (110)	37-41 Precision orientation determination
		12.9-14.7% Al		As in Fig. 6	Approx. (122) 20° from (110)	37-40, 19 Precision orientation determination
Lithium	..	'Pure'	BCC \rightarrow CPH The hexagonal layers are in faulty stacking sequence	$\{110\} \parallel \{00.1\}$ CPH $\langle 111 \rangle$ 3° from $\langle 11.0 \rangle$ CPH	(144), which is $10\frac{1}{2}^\circ$ from (110)	42, 15, 43
7 Au-Cd	..	47.5 at. % Cd	BCC \rightarrow Orthorhombic β'	$\{011\}\beta \parallel \{001\}\beta'$ $[111]\beta \parallel [110]\beta'$	(133)	9
	..	'Pure'	BCC \rightarrow CPH	$\{110\} \parallel \{00.1\}$ $\langle 111 \rangle$ 0° to 2° from $\langle 11.0 \rangle$	Unknown	44
Cobalt	'Pure'	FCC \rightarrow CPH	$\{111\} \parallel \{00.1\}$ $\langle 110 \rangle \parallel \langle 11.0 \rangle$	(111)	44, 45 Habit plane is from unpublished work by the authors
In-Tl	20.75 at. % Tl	FCC \rightarrow FCT	As in Fig. 10	(110)	7 Analogous transformations probably occur in the systems In-Cd ⁴⁶ , Cu-Mn ⁴⁷ and Cr-Mn ⁴⁸

* The directions lie in the parallel planes

Subsequent determination of the orientation relationship, habit planes, and change in shape of the transformed volume have shown quite clearly that this transformation does not occur in this way. However, it is interesting to note that the correspondence between lattice positions before and after transformation that was suggested by Bain has been retained as a common feature of all mechanisms that have since been suggested for the transformation.

The next stage in the development of theories of atom movements was the determination of the orientation relationships that exist between parent and martensitic phases, and the postulation of mechanisms which attempted to produce the correct martensite

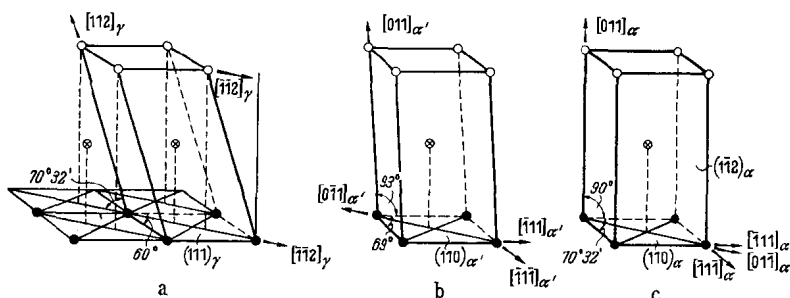


Figure 2. Atom movements postulated by Kurdjumov and Sachs for the transformation from A austenite (γ), to B tetragonal martensite (α'), and C to cubic ferrite (α). The bases of the cells indicated are (111) in γ and (110) in α' and α .

structures and the correct orientation relationships to the parent lattice. The available data on orientation relationships in martensite transformations are summarized in Table I.

KURDJUMOW and SACHS^{31, 49} determined the orientation relationship between austenite and martensite in 1.4 per cent carbon steel, and proposed a transformation mechanism to account for this relationship. They proposed that martensite is formed by the two consecutive shears $(111)_A [\bar{1}\bar{1}2]_A$ and $(1\bar{1}2)_M [\bar{1}11]_M$. The movement of atoms produced by these shears is illustrated in Figure 2. This mechanism leads to the correct orientation relationship and to approximately the correct structure.* However, as was found subsequently, it is not consistent with the observed habit plane and relief effects on polished surfaces.

A slightly different orientation relationship was found in a 70 : 30 iron-nickel alloy by NISHIYAMA,³³ who accordingly proposed a somewhat different transformation mechanism. Nishiyama proposed that

* Certain readjustments of atom positions must occur before the new structure has the correct dimensions. Moreover, the later geometrical analyses of this transformation by JASWON and WHEELER²¹ and by BOWLES⁵⁰ show that these two shears are not capable of producing an exactly body-centred cubic structure.

this transformation occurs by means of a single shear of $19^{\circ} 28'$ on $(111)_A [\bar{1}\bar{1}2]_A$. This is the same as the first step of the Kurdjumow-Sachs mechanism. The Kurdjumow-Sachs second shear, although it would produce approximately the desired cubic structure, is not permissible in this case since in the Nishiyama relationship the $[\bar{1}\bar{1}2]_A$ direction is parallel to the $[1\bar{1}0]_M$ direction. Thus the enlargement of the basal angle of *Figure 2* from 60° to $70^{\circ} 38'$, which was accomplished by the second shear in the Kurdjumow-Sachs mechanism, must be

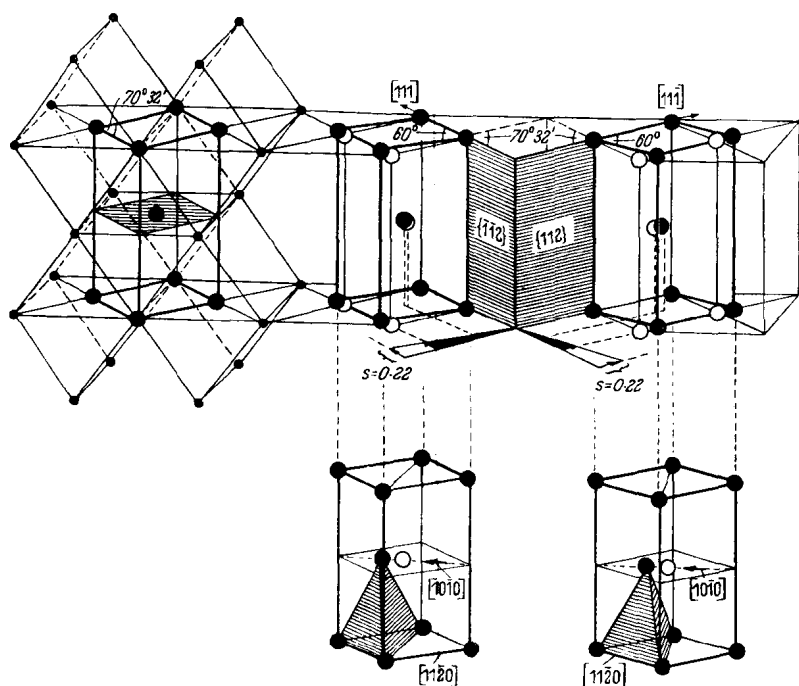


Figure 3. Atom movements postulated by Burgers for the body-centred cubic to close-packed hexagonal transformation in zirconium. On the left are body-centred cubic cells, and in heavy lines a cell having $(\bar{1}01)_{bcc}$ as a base and $\{112\}_{bcc}$ as vertical sides, the latter serving as shear planes when the two hexagonal cells at the lower right are produced

accomplished by 'readjustments' in the Nishiyama mechanism. This mechanism also leads to the observed orientation relationship but again is not consistent with the observed habit plane and relief effects.

From a study of the orientation relationship in zirconium (*Table I*) BURGERS⁴⁴ proposed that the martensite transformation of body-centred cubic (bcc) to close-packed hexagonal (cph) occurs by a heterogeneous shear on the system $(112)_{bcc} [\bar{1}\bar{1}\bar{1}]_{bcc}$. This distortion is illustrated in *Figure 3*. The crystallographic principle underlying this proposal is that the configuration of atoms in the (112) plane of a

body-centred cubic structure is exactly the same as that in the (10 $\bar{1}$ 0) plane of a close-packed hexagonal structure based on the same atom radius. The pattern on both these planes is a rectangle, $d \times (2\sqrt{2}/\sqrt{3})d$, where d is the atom diameter. Thus a hexagonal close-packed structure can be built up from a body-centred cubic structure by displacing the (112) planes relative to each other. It should be noted, however, that the array on the (110)_{bcc} planes (see *Figure 3*)

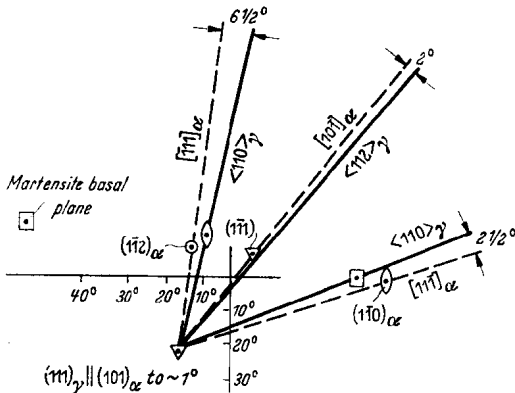


Figure 4. Orientation relationships in the martensite transformation, γ to α , in iron-0.8 per cent carbon-22 per cent nickel. Gnomonic projection with the plane of the martensite plate as the plane of projection (Greninger and Troiano)

cannot be converted into an exact hexagonal array merely by the pure shear proposed by Burgers since the spacings of the $(112)_{\text{bcc}}$ planes and $(10\bar{1}0)_{\text{eph}}$ planes into which they transform are not equal. Nevertheless this mechanism leads to approximately the observed orientation relationship.

BURGERS also proposed⁴⁴ a mechanism for the martensite transformation from the face-centred cubic structure to the

close-packed hexagonal structure that occurs in cobalt. In this transformation, since the (111) planes in the face-centred cubic phase and the basal planes in the close-packed hexagonal phase have the same configuration of atoms, the change in structure could be produced by displacement of the (111) planes relative to each other. Burgers proposed that this transformation occurs by a shear on the system (111) $[\bar{1}\bar{1}2]$ as illustrated in *Figure 7*. This shear only occurs on alternate planes and so converts the **ABCABC** . . . sequence of (111) planes in the face-centred cubic structure with the **ABABAB** . . . sequence of basal planes in the close-packed hexagonal structure. This mechanism is consistent with the observed orientation relationship.

With the exception of the mechanism proposed for the face-centred cubic \rightarrow close-packed hexagonal transformation, none of these transformation mechanisms has proved to be capable of explaining the observed habit planes and relief effects. Prior to the determination of the martensite habit planes it was generally believed that these planes would be the operative shear planes in the various mechanisms described above. However, the experimental determinations did not substantiate this prediction. In general, martensite plates do not form