OVERVIEW

Invariant-Plane and Invariant-Line Deformation Criteria and Their Application to Interface Crystallography*

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Simple geometrical approaches to interface crystallography based on the invariant-plane and invariant-line deformation criteria have been overviewed. Using matrix algebra, the mathematical implications of these criteria are first discussed. To elucidate the physical meaning and the applicability of the invariant-plane deformation criterion, the re-formulation procedure of the phenomenological crystallographic theories of martensitic transformations using the infinitesimal-deformation approach is surveyed. As the application of the invariant-line deformation criterion, epitaxial relationships between deposit thin-film crystals and substrate crystals are analyzed. It has been demonstrated that these simple geometrical criteria are very useful in explaining various features of the crystallography of interfaces.

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I. Introduction

When two different crystals meet at a sharp interface, the two crystals often exhibit a certain crystallographic orientation relationship. For example, at interfaces between f.c.c. and b.c.c. crystals, such well-known orientation relationships as Kurdjumov-Sachs (KS)⁽¹⁾, Nishiyama-Wassermann (NW)⁽²⁾⁽³⁾ and Pitsch⁽⁴⁾ are often observed. However, it is not necessarily easy to understand the reason why a particular relationship is realized in a given system.

It is often said that there are two kinds of inter-crystalline interfaces depending on whether or not their crystallography can be explained in a geometrical manner. In this overview, only such interfaces as can be understood geometrically will be discussed. Moreover, the main focus will be placed on the interfaces to which invariant-plane and invariant-line criteria are applicable. As will be demonstrated below, analyses based on these criteria are simple and straightforward. Nevertheless, many features on the crystallography and morphology of various interfaces can be explained clearly. For more involved geometrical analyses which will not be covered in this overview, the readers should consult excellent review articles available in the literature⁽⁵⁾⁻⁽⁹⁾.

II. Geometrically Favorable Interfaces

In general, a vector x in the three-dimensional space is transformed into another vector y by a 3×3 linear transformation matrix A as

$$y = Ax. \quad (1)$$

Let us consider two crystal lattices 1 and 2. When A is regarded as a matrix to transform lattice 1 to lattice 2, Bollmann's O-lattice theory⁽¹⁰⁾, one of the well-known geometrical theories for crystal boundaries and interfaces, states that a so-called O-lattice vector $X^{(O)}$ can be defined by the following equation:

$$(I - A^{-1})X^{(O)} = b^{(L)}, \quad (2)$$

where I is a 3×3 unit matrix, A^{-1} is the reciprocal matrix of A and $b^{(L)}$ is an arbitrary lattice translation vector of lattice 1. Equation (2) constitutes three simultaneous equations to solve the three unknown components of $X^{(O)}$.

It is generally considered⁽⁶⁾⁻⁽⁸⁾ that better geometrical lattice matching at an interface between two different lattice planes is attained as the absolute magnitude of the determinant of $I - A^{-1}$, i.e. $|\det(I - A^{-1})|$, becomes smaller. Since this matrix is a 3×3 matrix, its rank r can be 0, 1, 2 or 3. When $r=3$, $X^{(O)}$ can be determined uniquely for given $b^{(L)}$ and, accordingly, by choosing various $b^{(L)}$ vectors, a set of so-called O-lattice points can be defined. On the other hand, when $r \neq 3$, the three simultaneous eq. (2) are not independent of each other and $|\det(I - A^{-1})|$ takes its minimum value of zero, as is well known in matrix algebra. If this is the case, therefore, the optimum geometrical matching between the two lattices can be attained. In this overview, geometrical theories and their physical implications for such special cases of

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$r \neq 3$ will be reviewed and the theories will be applied to discuss the crystallography and morphology of two crystals which meet at an interface. It should be mentioned here that $r=0$ is realized only when $A=I$, resulting in the perfect matching of the two crystals at any interface orientation (A in this case represents an “invariant-body” deformation). Therefore, this case will not be treated below and only the cases for $r=1$ and 2 will be discussed.

III. Interfaces Described by $r=1$ —Invariant Plane Deformation

When $r=1$, only one of the three simultaneous eq. (2) is independent and, therefore, the solutions for $X^{(0)}$ represent a set of planes (O-planes)⁽¹⁰⁾. Let us choose an (x_1, x_2, x_3) right-handed orthonormal coordinate system as a reference system. It is easily understood that the rank of the matrix $I-A^{-1}$ is one when the matrix A takes the following form.

$$A = \begin{pmatrix} 1 & 0 & A_{13} \\ 0 & 1 & A_{23} \\ 0 & 0 & 1+A_{33} \end{pmatrix}. \quad (3)$$

As shown in Fig. 1, this matrix expresses an invariant-plane deformation with the x_3 -axis perpendicular to the invariant plane (or the x_3 -plane parallel to the invariant plane). A_{13} and A_{23} are the shear components parallel to the invariant plane and A_{33} is the dilatational component perpendicular to the invariant plane. For a special case of $A_{33}=0$, the deformation becomes a simple shear. In this overview, the word “invariant” is used when both “unstrained” and “unrotated” conditions are satisfied simultaneously.

The so-called phenomenological crystallographic theo-

ries of martensitic transformations⁽¹¹⁾⁽¹²⁾ are among the most well-known geometrical theories based on the invariant-plane deformation. In these theories, the crystallography and morphology of a plate-shaped martensite are considered to be determined so as to make the interface plane (the habit plane) between the martensite and the parent phase invariant.

In general, the unstrained-plane deformation condition, a necessary condition for the invariant-plane deformation, cannot be satisfied by the operation of homogeneous lattice deformation B (for example, the well-known Bain deformation) alone, which transforms a parent lattice into a martensite lattice. Therefore, in the light of experimental observation that martensites have internal structures caused by slip or twinning, additional deformation (so-called lattice-invariant deformation) P consisting of a simple shear by slip or twinning is introduced. Moreover, to meet the unrotated-plane condition or to make the habit plane unrotated, rigid-body rotation R is also considered to be operative. The total shape deformation associated with the martensitic transformation is thus expressed by the multiplication of these three matrices as

$$A = RBP. \quad (4)$$

Here, the matrix B can be found from the lattice constants of the parent and martensite phases together with the assignment of a proper lattice correspondence between the two phases. Although a particular slip system (or a twinning system) is assumed to describe P , the amount of slip (or the volume fraction of one of the two twin-related crystals) is an unknown quantity. In the phenomenological crystallographic theories, the unknown quantity and R are determined so as to make the matrix A express an invariant-plane deformation of the form of eq. (3). As a result, such crystallographic solutions as habit-plane orientation, the amount of slip (or the twin volume fraction), the orientation relationship between the parent and martensite phases, etc. can be obtained.

The phenomenological crystallographic theories have already been formulated completely. No further fundamental theoretical development or refinement can be expected. However, since the theories are described by the finite-deformation (FD) approach, the mathematical procedure to obtain crystallographic and morphological solutions is rather complicated and computer calculation is almost certainly inevitable. Therefore, the relationship between input data and output solutions can hardly be understood analytically.

Keeping such complications in mind, we have attempted to simplify the phenomenological theories by reformulating them using the infinitesimal-deformation (ID) approach⁽¹³⁾⁽¹⁴⁾. In the ID approach, second- and higher-order terms of strain and rotation components are neglected by assuming that these components are much smaller than unity. In other words, the ID analysis is an approximation of the FD analysis. After the approximation, an ID matrix is obtained by subtracting a unit

Fig. 1 Invariant-plane deformation with the x_3 -plane parallel to the invariant plane.

matrix from the corresponding FD matrix⁽¹³⁾⁽¹⁴⁾. With the above procedure, the sequential occurrence of deformations and rotations is expressed by the summation (not by the multiplication) of ID matrices. Although matrix multiplication is in general uncommutable, summation is commutable. Therefore, the ID analysis is much simpler than the FD analysis. For example, eq. (4) can be rewritten on the basis of the ID approach as

$$\mathbf{A}^S = \mathbf{R}^S + \mathbf{B}^S + \mathbf{P}^S, \quad (5)$$

where $\mathbf{A}^S$, $\mathbf{R}^S$, $\mathbf{B}^S$ and $\mathbf{P}^S$ are the ID matrices corresponding respectively to $\mathbf{A}$, $\mathbf{R}$, $\mathbf{B}$ and $\mathbf{P}$ in eq. (4).

When $\mathbf{A}$ is given by eq. (3), $\mathbf{A}^S$ to describe the invariant-plane deformation in the ID approach becomes

$$\begin{aligned} \mathbf{A}^S &= \begin{pmatrix} 0 & 0 & A_{13} \\ 0 & 0 & A_{23} \\ 0 & 0 & A_{33} \end{pmatrix} \\ &= \mathbf{A}^S(\text{sym.}) + \mathbf{A}^S(\text{anti.}) \\ &= \begin{pmatrix} 0 & 0 & A_{13}/2 \\ 0 & 0 & A_{23}/2 \\ A_{13}/2 & A_{23}/2 & A_{33} \end{pmatrix} \\ &\quad + \begin{pmatrix} 0 & 0 & A_{13}/2 \\ 0 & 0 & A_{23}/2 \\ -A_{13}/2 & -A_{23}/2 & 0 \end{pmatrix}. \end{aligned} \quad (6)$$

Here, the first term, the symmetric matrix $\mathbf{A}^S(\text{sym.})$, represents strain components and the second term, the antisymmetric matrix $\mathbf{A}^S(\text{anti.})$ represents rotation components⁽¹³⁾⁽¹⁴⁾. As can be seen from eq. (6), the condition for the x_3 -plane to be the unstrained plane is satisfied when the strain components 11, 12 and 22 of $\mathbf{A}^S(\text{sym.})$ vanish. Equivalently, the diagonalization of $\mathbf{A}^S(\text{sym.})$ reveals that the unstrained-plane condition is satisfied when one of the three principal strains is zero and the other two be respectively positive and negative⁽¹³⁾. A corresponding unstrained-plane condition has also been discussed in the FD-based phenomenological theories⁽¹¹⁾⁽¹⁵⁾⁽¹⁶⁾.

Using ID-based Eshelby's ellipsoidal inclusion problem in elasticity⁽¹⁷⁾, it has been shown⁽¹⁸⁾⁽¹⁹⁾ that if the unstrained-plane condition is satisfied, no elastic strain energy is created upon the formation of plate-shaped martensites. In other words, the predicted crystallography and morphology of martensites by the phenomenological theories are the most ideal ones from the viewpoint of elastic strain energy minimization. This is the reason why an invariant-plane interface described by $r=1$ is not only geometrically but also physically favorable.

As mentioned earlier, the FD-based phenomenological theories, though ingeniously formulated, have a drawback, if any, that the solutions for habit-plane orientation, orientation relationship, etc. are obtained only numerically by computer calculation. On the other hand, the ID approach makes it possible to obtain these solutions analytically without using a computer.

To exemplify the ID-based phenomenological analysis and to enable direct comparison with the original FD analysis, let us consider an f.c.c. to b.c.c. martensitic transformation⁽¹³⁾. As a lattice correspondence, the usual Bain correspondence shown in Fig. 2 will be adopted. That is, the correspondence between $(h k l)_f$ and $(p q r)_b$ planes (The subscripts f and b denote f.c.c. and b.c.c. crystals, respectively.) is written as

$$(p q r)_b = (h k l)_f \begin{pmatrix} 1/2 & 1/2 & 0 \\ -1/2 & 1/2 & 0 \\ 0 & 0 & 1 \end{pmatrix} \quad (7)$$

and the directional correspondence between $[a b c]_f$ and $[u v w]_b$ directions is

$$\begin{bmatrix} u \\ v \\ w \end{bmatrix}_b = \begin{pmatrix} 1 & -1 & 0 \\ 1 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix} \begin{bmatrix} a \\ b \\ c \end{bmatrix}_f. \quad (8)$$

The lattice deformation (the Bain deformation) matrix $\mathbf{B}^S$ is then written on the basis of the f.c.c. coordinate system (the f system), i.e. $\mathbf{x}_1^{(f)} \parallel [100]_f$, $\mathbf{x}_2^{(f)} \parallel [010]_f$ and $\mathbf{x}_3^{(f)} \parallel [001]_f$, as

$$\mathbf{B}^S = \begin{pmatrix} \varepsilon_1 & 0 & 0 \\ 0 & \varepsilon_1 & 0 \\ 0 & 0 & \varepsilon_2 \end{pmatrix}_f. \quad (9)$$

Here, using the lattice parameters a_f and a_b of the f.c.c. matrix and the b.c.c. martensite, the principal Bain strains are obtained as $\varepsilon_1 = \sqrt{2}(a_b/a_f) - 1$ and $\varepsilon_2 = (a_b/a_f) - 1$, as can be understood from Fig. 2. For the $\gamma \rightarrow \alpha$ transformation in ferrous alloys, ε_1 and ε_2 are about 0.12 and -0.20 , respectively.

As the lattice-invariant deformation $\mathbf{P}^S$, the $(011)_f$ $[\bar{0}\bar{1}1]_f$ slip system will be assumed, as adopted by the original phenomenological theories. Then, the matrix $\mathbf{P}^S$ on the f system can be written as

$$\mathbf{P}^S = \begin{pmatrix} 0 & 0 & 0 \\ 0 & -m/2 & -m/2 \\ 0 & m/2 & m/2 \end{pmatrix}_f, \quad (10)$$

where m is the unknown magnitude of the slip shear. The

Fig. 2 The Bain correspondence between f.c.c. and b.c.c. crystals.

rigid-body rotation R^S in the ID analysis is expressed in general form as an antisymmetric matrix:

$$R^S = \begin{pmatrix} 0 & -\omega_3 & \omega_2 \\ \omega_3 & 0 & -\omega_1 \\ -\omega_2 & \omega_1 & 0 \end{pmatrix}_f, \quad (11)$$

where ω_i ($i=1, 2$ or 3) is an unknown angle of rotation about the $x_i^{(f)}$ -axis.

From eqs. (5) and (9) to (11), the total shape deformation A^S can be written as

$$A^S = \begin{pmatrix} \varepsilon_1 & -\omega_3 & \omega_2 \\ \omega_3 & \varepsilon_1 - m/2 & -\omega_1 - m/2 \\ -\omega_2 & \omega_1 + m/2 & \varepsilon_2 + m/2 \end{pmatrix}_f. \quad (12)$$

A necessary condition for A^S to describe the invariant-plane deformation is that A^S (sym.), i.e.

$$A^S(\text{sym.}) = \begin{pmatrix} \varepsilon_1 & 0 & 0 \\ 0 & \varepsilon_1 - m/2 & 0 \\ 0 & 0 & \varepsilon_2 + m/2 \end{pmatrix}_f \quad (13)$$

must represent an unstrained-plane deformation. As mentioned earlier, for this to be the case, one of the principal strains should be zero. Since ε_1 is not zero, either $\varepsilon_1 - m/2 = 0$ or $\varepsilon_2 + m/2 = 0$ must be satisfied. Let us consider the former case in the following. (The latter case can be treated in a similar manner⁽¹³⁾.) Insertion of $\varepsilon_1 = m/2$ into eq. (13) yields

$$A^S(\text{sym.}) = \begin{pmatrix} \varepsilon_1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & \varepsilon_1 + \varepsilon_2 \end{pmatrix}_f \quad (14)$$

Here, a new n coordinate system with $x_1^{(n)} \parallel [\cos \theta \times \cos \phi, \cos \theta \sin \phi, -\sin \theta]_f$, $x_2^{(n)} \parallel [-\sin \phi, \cos \phi, 0]_f$ and $x_3^{(n)} \parallel [\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta]_f$, where θ and ϕ are unknown angles to be determined, will be chosen so that the $x_3^{(n)}$ -plane becomes parallel to the habit plane (the invariant plane). Then, $A^S(\text{sym.})$ expressed on this system must have the form of the first term in the right-hand side of eq. (6). Conducting the transformation of coordinate systems for strain tensors, we find that the above requirement leads to $\sin \theta = (-\varepsilon_1 / \varepsilon_2)^{1/2}$ as one of crystallographically equivalent solutions⁽¹³⁾. From this, the orientation of the habit plane normal, i.e. the $x_3^{(n)}$ -axis, is obtained in the f system as

$$x_3^{(n)} \parallel [(-\varepsilon_1 / \varepsilon_2)^{1/2}, 0, \{(\varepsilon_1 + \varepsilon_2) / \varepsilon_2\}^{1/2}]_f \quad (15)$$

We have so far used only the unstrained-plane condition by taking into account the symmetric component $A^S(\text{sym.})$ of A^S . In order for the habit plane to be an invariant plane, this plane must also be unrotated.

Since $\varepsilon_1 = m/2$ has been chosen for the present discussion, eq. (12) becomes

$$A^S = \begin{pmatrix} \varepsilon_1 & -\omega_3 & \omega_2 \\ \omega_3 & 0 & -\varepsilon_1 - \omega_1 \\ -\omega_2 & \varepsilon_1 + \omega_1 & \varepsilon_1 + \varepsilon_2 \end{pmatrix}_f. \quad (16)$$

To find ω_i 's, let us choose two non-parallel vectors V_1 and V_2 on the habit plane as follows:

$$V_1 = [\{(\varepsilon_1 + \varepsilon_2) / \varepsilon_2\}^{1/2}, 0, -(-\varepsilon_1 / \varepsilon_2)^{1/2}]_f \\ V_2 = [0, 1, 0]_f$$

These vectors on the habit plane should remain unchanged by the operation of the invariant-plane deformation. In the ID analysis, this is equivalent to saying that $A^S V_1 = A^S V_2 = [0, 0, 0]$ must be satisfied⁽¹³⁾. From these conditions, the total deformation A^S can be found on the f and n coordinate systems as follows.

$$A^S = \begin{pmatrix} \varepsilon_1 & 0 & \varepsilon_1 \{-(\varepsilon_1 + \varepsilon_2) / \varepsilon_1\}^{1/2} \\ 0 & 0 & 0 \\ -\varepsilon_1 \{-(\varepsilon_1 + \varepsilon_2) / \varepsilon_1\}^{1/2} & 0 & \varepsilon_1 + \varepsilon_2 \end{pmatrix}_f \quad (17)$$

$$A^S = \begin{pmatrix} 0 & 0 & 2\varepsilon_1 \{-(\varepsilon_1 + \varepsilon_2) / \varepsilon_1\}^{1/2} \\ 0 & 0 & 0 \\ 0 & 0 & 2\varepsilon_1 + 2\varepsilon_2 \end{pmatrix}_n \quad (18)$$

Equation (18) shows that the total shape change on the formation of a plate-shaped martensite consists of the shear component $2\varepsilon_1 \{-(\varepsilon_1 + \varepsilon_2) / \varepsilon_1\}^{1/2}$ parallel to the habit plane and the dilatational component $2\varepsilon_1 + \varepsilon_2$ perpendicular to the habit plane. As expected, eq. (18) has the form of eq. (6).

Table 1 summarizes a set of ID-based analytical crystallographic solutions thus obtained⁽¹³⁾. The case for the $(011)_f$ $[0\bar{1}1]_f$ twinning as a lattice invariant shear deformation has also been analyzed⁽¹³⁾ and the obtained solutions are also shown in Table 1. These simple analytical solutions are useful and advantageous since they enable us to know easily how the martensite crystallography depends on the input values of ε_1 and ε_2 .

Although the above-mentioned ID-based theory is an approximation of the FD-based phenomenological theories, the ID solutions must become identical to the corresponding exact FD solutions at the limit of $\varepsilon_1 \rightarrow 0$ and $\varepsilon_2 \rightarrow 0$. In general, however, ID solutions should contain relative errors of the order of magnitude $|\varepsilon_1|$ or $|\varepsilon_2|$. If this order is denoted by Δ , since $\Delta \sim 0.1$ for the

Table 1 Crystallographic solutions for f.c.c. to b.c.c. martensitic transformations obtained by the ID analysis.

Habit plane	$\{(-\varepsilon_1 / \varepsilon_2)^{1/2}, 0, \{(\varepsilon_1 + \varepsilon_2) / \varepsilon_2\}^{1/2}\}_f$	
Total shape deformation		
shear component	$2\varepsilon_1 \{-(\varepsilon_1 + \varepsilon_2) / \varepsilon_1\}^{1/2}$	
dilatational component	$2\varepsilon_1 + \varepsilon_2$	
Lattice invariant deformation	Slip	Twinning
Amount of slip	$2\varepsilon_1$	—
Twin volume ratio	—	$ \varepsilon_1 : \varepsilon_2 $
Orientation relationship (tilt angles)		
[100] _f	$\{-(\varepsilon_1(\varepsilon_1 + \varepsilon_2))^{1/2} / \varepsilon_1\}$	$\{-(\varepsilon_1(\varepsilon_1 + \varepsilon_2))^{1/2} / \varepsilon_1\}$
[010] _f	$ \varepsilon_1 $	$ \varepsilon_1 $
[001] _f	$(-\varepsilon_1 \varepsilon_2)^{1/2}$	$(-\varepsilon_1 \varepsilon_2)^{1/2}$
	Twin I	Twin II
	$\{-(\varepsilon_1(\varepsilon_1 + \varepsilon_2))^{1/2} / \varepsilon_1\}$	$\{-(\varepsilon_1(\varepsilon_1 + \varepsilon_2))^{1/2} / \varepsilon_1\}$
	$ \varepsilon_1 $	$ \varepsilon_2 $
	$(-\varepsilon_1 \varepsilon_2)^{1/2}$	$\{-(\varepsilon_1(\varepsilon_1 + \varepsilon_2) + \varepsilon_2^2)\}^{1/2}$

$\gamma \rightarrow \alpha$ martensitic transformation in ferrous alloys, the approximate ID solutions for the habit-plane orientation, for example, may be about 10° (~ 0.1 rad) different from the corresponding exact FD solutions. On the other hand, the difference is much smaller, at most 1° , for the cubic to tetragonal transformation in zirconia alloys with $\Delta \sim 0.01$ ⁽¹³⁾.

Let us now consider a special case of the ID solutions. It is found from Table 1 for the twinned martensite that when the volume change associated with the transformation is zero, or $2\varepsilon_1 + \varepsilon_2 = 0$, the habit plane orientation becomes (101) and the twin volume ratio becomes 1:2. Figure 3(a) shows the morphology of two crystallographically equivalent plate-shaped martensites satisfying this special case. When these two martensites are formed alternately and side by side, a structure shown in Fig. 3(b) is realized. This space-filling structure is favorable from the viewpoint of elastic strain energy minimization, as mentioned previously. In fact, this morphology has been commonly observed not only in martensites but also in the domain structure of ferroelectric materials⁽²⁰⁾. In particular, this structure in zirconia alloys is well-known as the herringbone structure⁽²¹⁾⁻⁽²³⁾.

As demonstrated above, when the phenomenological crystallographic theories are described by the ID approach, not only the physical meaning of the theories becomes very clear but also simple analytical crystallographic solutions can be obtained. Although approximations, these solutions are often accurate enough for tenable comparison with experimental results. Moreover, they are very useful in finding the systematic variation of martensite crystallography as a function of input data. Using the ID approach, we have also derived analytical solutions for more complex crystallography associated with cubic to monoclinic martensitic transformations such as those in TiNi and zirconia alloys⁽¹⁴⁾. Satisfactory agreement between the ID and FD solutions has also been found even in these cases.

Fig. 3 The morphology of twinned martensites for a special case of $2\varepsilon_1 + \varepsilon_2 = 0$ obtained from the ID solutions in Table 1. (a) two crystallographically equivalent variants with a common (101) habit plane. (b) alternate and side-by-side arrangement of the two variants to form a space-filling structure.

IV. Interfaces Described by $r=2$ —Invariant Line Deformation

When $r=2$, two of the three simultaneous eq. (2) are independent and, therefore, the solutions for $X^{(0)}$ represent a set of straight lines (O-lines)⁽¹⁰⁾. The rank of the matrix $I - A^{-1}$ is two when A takes the following form:

$$A = \begin{pmatrix} 1 & A_{12} & A_{13} \\ 0 & 1 + A_{22} & A_{23} \\ 0 & A_{32} & 1 + A_{33} \end{pmatrix}. \quad (19)$$

This matrix expresses an invariant-line deformation with the x_1 -axis parallel to the invariant line.

As one of the representative examples for the application of the invariant-line deformation criterion, epitaxial interfaces between deposit thin-film crystals and substrate crystals can be referred to. The epitaxial interfaces are geometrically different from the parent phase/martensite interfaces. As discussed in the previous section, the orientation of a martensite habit plane can be chosen appropriately from the whole three-dimensional space so as to satisfy the invariant-plane deformation condition. For the epitaxial interfaces, on the contrary, since the orientation of a substrate plane is predetermined, efforts to seek an invariant-plane interface will in general be fruitless. Therefore, as a compromise, we should look for an interface satisfying the invariant-line deformation condition. Since the geometry involved is the deposit and substrate planes at the interface, the problem becomes two-dimensional. Accordingly, a matrix A to describe the invariant-line deformation is written on a two-dimensional (x_1, x_2) reference coordinate system as a 2×2 matrix:

$$A = \begin{pmatrix} 1 & A_{12} \\ 0 & 1 + A_{22} \end{pmatrix} \quad (20)$$

with the rank of $I - A^{-1}$ being one. Needless to say, A_{12} represents the shear component parallel to the invariant line and A_{22} the dilatational component perpendicular to the invariant line. For such a two-dimensional problem, even the FD analysis becomes very simple, as will be shown below.

By assuming a lattice correspondence between deposit and substrate crystals, the FD-based homogeneous lattice deformation matrix D to transform the atomic arrangement of the substrate plane into that of the deposit plane can be expressed as

$$D = \begin{pmatrix} 1 + \varepsilon_1 & 0 \\ 0 & 1 + \varepsilon_2 \end{pmatrix}_s \quad (21)$$

Here, the subscript s indicates that D is expressed on an appropriately chosen ($x_1^{(s)}, x_2^{(s)}$) substrate coordinate system (the s system). ε_1 and ε_2 are the principal strains which are determined by the lattice correspondence and the lattice parameters of the deposit and substrate

Fig. 4 Formation of an invariant line by the operation of (a) homogeneous deformation D and (b) rotation R .

crystals. On the s system shown in Fig. 4(a), the matrix D transforms a unit circle A (representing the substrate plane) into an ellipse B (deposit plane) with the aspect ratio $(1 + \varepsilon_2)/(1 + \varepsilon_1)$. With this transformation a vector V_C on the circle changes into a vector V_E on the ellipse. If the ellipse is rotated with respect to the circle by θ about the origin, the two vectors coincide to be V_I . This V_I vector is an invariant vector since both its direction and length remain unchanged before and after the transformation. The matrix R to express the θ rotation is given in the FD representation by

$$R = \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix} \quad (22)$$

Therefore, the total deformation matrix A can be written as

$$A = RD. \quad (23)$$

Comparison of eq. (23) with eq. (4) reveals that the present invariant-line analysis of the epitaxial interfaces corresponds to a two-dimensional version of the phenomenological crystallographic theories. Since the invariant-line deformation condition is satisfied by the operation of only D and R , the lattice-invariant deformation P is not needed for the present two-dimensional problem.

Similar to the previously-mentioned condition for principal strains to realize the unstrained-plane deformation, the condition for the unstrained-line deformation exists, i.e. ε_1 and ε_2 have different signs, or $\varepsilon_1 \varepsilon_2 < 0$. When this condition is satisfied, the rotation angle θ to realize the invariant-line deformation condition can be easily obtained from $\det(I - A^{-1}) = 0$ and eqs. (21) to (23) as⁽²⁴⁾

$$\cos \theta = (1 + \eta_1 \eta_2) / (\eta_1 + \eta_2), \quad (24)$$

where, $\eta_i = 1 + \varepsilon_i$ ($i = 1, 2$). Then, the normalized invariant-line vector V_I becomes⁽²⁵⁾

$$V_I = [\pm \{(\eta_2^2 - 1)/(\eta_2^2 - \eta_1^2)\}^{1/2}, \pm \{(1 - \eta_1^2)/(\eta_2^2 - \eta_1^2)\}^{1/2}]_s. \quad (25)$$

Since D and R are now known, A in eq. (23) can be obtained. When another new $(x_1^{(i)}, x_2^{(i)})$ coordinate system (the i system) with the $x_1^{(i)}$ -axis parallel to V_I is used to

express A , we can easily obtain

$$A = \begin{pmatrix} 1 & \pm \{(1 - \eta_1^2)(\eta_2^2 - 1)\}^{1/2} \\ 0 & \eta_1 \eta_2 \end{pmatrix}_i \quad (26)$$

Equation (26) has the form of eq. (20), as it should.

To investigate the applicability of the invariant-line deformation condition to the epitaxial interfaces, we have used the Cu/Fe system for experiments, one as a substrate crystal and the other as a deposit crystal^{(25)–(31)}. Specimens of Cu or Fe single crystals for transmission electron microscopy (TEM) with various surface orientations were first prepared and they were used as substrates. Using a vacuum evaporation apparatus, thin films of the other crystal with average thickness of about 10–50 nm were deposited on the substrates. Epitaxial relationships on various substrate planes observed by TEM are summarized in Table 2. It is very interesting to find from Table 2 that f.c.c. and b.c.c. planes to form an interface are always related to each other by the Bain correspondence of eq. (7).

Once f.c.c./b.c.c. interface plane orientations are found, the matrix D in eq. (21) can be obtained as follows. For the demonstration, let us consider the case of the f.c.c. substrate/b.c.c. deposit system. From eq. (7), we find that the $(h \ k \ l)_f$ plane corresponds to the $([h-k]/2, [h+k]/2, l)_b$ plane, constituting the f.c.c./b.c.c. interface. On these two planes, two mutually perpendicular directions of principal strains, f_1 and f_2 (on the f.c.c. plane) and b_1 and b_2 (on the b.c.c. plane), can be found uniquely so that f_1 and b_1 as well as f_2 and b_2 are related to each other by the Bain correspondence⁽²⁵⁾. The indices of these principal directions are shown in Table 3.

Table 2 Orientations of epitaxial interfaces observed for the Cu–Fe system.

Deposits	Substrates	Deposits	Substrates
Cu(001)	Fe(001)	Cu(012)	Fe(111)
	Fe(011)		Fe(013)
Cu(011)	Fe(001)		Fe(114)
	Fe(112)	Cu(013)	Fe(012)
Cu(111)	Fe(011)		Fe(233)
Cu(112)	Fe(123)		Fe(116)
	Fe(012)	Cu(123)	Fe(122)
Cu(122)	Fe(134)		Fe(136)
	Fe(012)	Fe(001)	Cu(001)
Cu(113)	Fe(013)	Fe(011)	Cu(111)
	Fe(112)	Fe(111)	Cu(012)
		Fe(112)	Cu(011)

Table 3 The Bain correspondences of f.c.c. and b.c.c. planes and principal directions forming an interface.

	f.c.c.		b.c.c.
Interface orientation	$(h, k, l)_f$	$\leftrightarrow$	$([h-k]/2, [h+k]/2, l)_b$
Principal directions	$f_1 = [hl, kl, -(h^2 + k^2)]_f$	$\leftrightarrow$	$b_1 = [(h-k)l, (h+k)l, -(h^2 + k^2)]_b$
	$f_2 = [-k, h, 0]_f$	$\leftrightarrow$	$b_2 = [-h-k, h-k, 0]_b$

In the present analysis, f_1 and f_2 thus determined should be regarded respectively parallel to $x_1^{(s)}$ - and $x_2^{(s)}$ -axes of the s system in Fig. 4. Then, the components of D can be expressed as

$$1 + \varepsilon_1 = \eta_1 = (|b_1|a_b)/(|f_1|a_f), \quad (27)$$

$$1 + \varepsilon_2 = \eta_2 = (|b_2|a_b)/(|f_2|a_f), \quad (28)$$

where a_b and a_f are the lattice parameters of b.c.c. and f.c.c. crystals. By knowing the values of eqs. (27) and (28), the invariant-line condition can be obtained from eqs. (24) and (25)[†].

As an example, let us analyze the epitaxy of an Fe film on the $(111)_f$ plane of Cu. From Table 3, the $(111)_f$ plane is found to correspond to the $(011)_b$ plane by the Bain correspondence. The principal directions are also found to be $f_1 = [11\bar{2}]_f$, $f_2 = [\bar{1}10]_f$, $b_1 = [02\bar{2}]_b$ and $b_2 = [\bar{2}00]_b$. Therefore, from eqs. (27) and (28), we have $\eta_1 = (2/\sqrt{3})a_b/a_f$ and $\eta_2 = \sqrt{2}a_b/a_f$. Insertion of these into eq. (24) leads to

$$\cos \theta = \{1 + (2\sqrt{2}/\sqrt{3})\rho^2\} / \{(\sqrt{2} + 2/\sqrt{3})\rho\}, \quad (29)$$

with $\rho = a_b/a_f$. As is well known, the exact KS and NW relationships are realized when $\theta = 5.26^\circ$ and 0° , respectively. These relationships are shown in Fig. 5. For the Cu/Fe system, ρ becomes 0.792 ($a_f = 0.361$ nm for Cu and $a_b = 0.286$ nm for Fe). Therefore, as an epitaxial relationship on the Cu $(111)_f$ //Fe $(011)_b$ interface, $\theta = 5.75^\circ$ is predicted from eq. (29). The fact that this θ value is very close to that of the KS relationship means that the present invariant-line analysis predicts essentially the KS relationship. Indeed, the KS relationship has been observed experimentally for the Cu/Fe system⁽²⁶⁾⁽²⁷⁾.

It is worth mentioning here that eq. (29) can be used to predict epitaxial relationships on the $(111)_f/(011)_b$ interfaces of various other f.c.c./b.c.c. combinations. Kato⁽³²⁾ has shown that the predicted epitaxial relationships for various f.c.c./b.c.c. metal combinations are

Fig. 5 (a) KS and (b) NW orientation relationships. $\circ$ f.c.c. atoms, $\bullet$ b.c.c. atoms.

[†] If ε_1 and ε_2 have the same sign, the invariant-line condition cannot be satisfied, as mentioned earlier. In this case, $\theta = 0^\circ$ is conventionally assumed to predict an epitaxial relationship for the later discussion.

not only in excellent agreement with experimental results available in literature but also they are almost the same as the predicted epitaxial relationships derived from more involved analyses based on the O-lattice theory⁽⁶⁾, moiré-pattern analysis⁽³³⁾ and computer calculation of interfacial energies⁽³⁴⁾.

By combining the Bain correspondence and the invariant-line criterion, epitaxies on other substrate plane orientations can also be predicted easily. In fact, we have been successful in predicting and explaining the observed epitaxial relationships listed in Table 2⁽²⁵⁾⁻⁽³¹⁾.

It should be noted here that given a substrate plane orientation other than (111) , the Bain lattice correspondence of eq. (7) predicts more than one candidate deposit planes. Consequently, more than one epitaxial relationships are predicted. Therefore, it becomes necessary to have other criteria to judge which of the predicted epitaxial relationships is most favorable. One criterion can be taken from the invariant-line condition per se. It has been suggested⁽²⁵⁾⁽³²⁾ that an epitaxy which can satisfy the invariant-line condition is more favorable than an epitaxy which cannot. However, in case when this criterion still fails to uniquely select a most favorable epitaxy, another criterion becomes necessary. For this purpose, we have defined the following parameter M which is approximately proportional to elastic strain energy due to the misfit between deposit and substrate lattice planes⁽²⁸⁾⁻⁽³²⁾⁽³⁵⁾:

$$M = \varepsilon_1^2 + \varepsilon_2^2 + (2/3)\varepsilon_1\varepsilon_2. \quad (29)$$

The proposed criterion is that the smaller the M parameter value, the more favorable an epitaxial relationship becomes. These new criteria have been applied successfully to explain epitaxial relationships observed (i) on the Cu/Fe system⁽²⁸⁾⁻⁽³²⁾, and (ii) at interfaces between various cubic deposit crystals and (001) cubic substrate crystals⁽³⁵⁾.

As mentioned above, by taking three independent factors into account, i.e. the Bain correspondence, the invariant-line criterion and the M parameter, epitaxial relationships observed at various interfaces have been analyzed in a simple geometrical manner. Of the three factors, the applicability of the Bain correspondence is limited only to the f.c.c./b.c.c. interfaces with the one to one correspondence between atoms in the two crystal planes. For an interface with other combinations of two crystals, the present geometrical analysis cannot be performed unless an appropriate lattice correspondence is found. Since infinite number of lattice correspondences can, in general, be considered for a given pair of deposit/substrate crystals, it is necessary to judge which correspondence is the most reasonable one. For this purpose, the idea of the "reduced superlattices" proposed by Zur and McGill⁽³⁶⁾ is useful. In fact, to explain the observed epitaxy of an Al thin film on the (001) plane of Si, this idea has been used very adroitly⁽³⁷⁾.

The epitaxies so far discussed are those observed on flat substrate crystals. It is interesting to know what happens if curved substrates are used instead. Wada *et al.*

examined this case by depositing Fe on the spherical substrates of Cu and Au⁽³⁸⁾⁻⁽⁴⁰⁾. For the spherical substrates of about 100 nm diameter, the tips of needle-like specimens for field ion microscopy (FIM) were used. Among various low-index substrate planes which were exposed on the spherical tip surfaces, strong tendencies towards the formation of Fe(011)_b on Cu(111)_f and Fe(100)_b on Au(100)_f were recognized through the FIM observation. Although details will not be repeated here, the invariant-line criterion as well as the *M* parameter analysis was found to be excellently applicable to explain the above and other crystallographic features of the deposited Fe films⁽³⁹⁾⁽⁴⁰⁾.

Aside from the epitaxy of two crystals, the invariant-line criterion has also been adopted to discuss the crystallography and morphology of various precipitate/matrix interfaces. For example, the growth direction of needle-like precipitates⁽⁴¹⁾⁻⁽⁴³⁾ and the formation of structural ledges at the precipitate/matrix interfaces⁽⁹⁾⁽⁴⁴⁾ have been explained satisfactorily.

V. Concluding Remarks

In these days, direct observation of atomic arrangements at interfaces and grain boundaries is possible through high-resolution electron microscopy. Advanced approaches combining high-speed computer technology and modern knowledge of solid-state physics and quantum chemistry are also being applied to analyze atomistic structures of interfaces. In comparison, such geometrical approaches as those overviewed above may appear too primitive and old-fashioned. It is in fact true that the structures of some interfaces are so complex that simple geometrical approaches become utterly powerless. However, having seen several examples of the excellent applicability of simple invariant-plane and invariant-line deformation criteria to the crystallography of interfaces, I am sure that one cannot help amazing and admiring the beauty and simplicity of these criteria that our predecessors elaborately constructed with a flash of genius.

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