Twinning in $\gamma'$ Cu–Al–Ni Martensite with Cu$_3$Ti Type Ordered Structure

By Kazuhiro Otsuka** and Ken'ichi Shimizu**

The {121}$\gamma'$ and {101}$\gamma'$ twinnings in $\gamma'$ Cu–Al–Ni martensite with Cu$_3$Ti type ordered structure, whose sublattice is HCP, have been analyzed by the theory of deformation twinning and the phenomenological theory of martensitic transformations. As a result the following conclusions have been reached:

1. The twinning elements of {121}$\gamma'$ twinning consistent with the phenomenological theory are as follows. $K_1 = (121)_{\gamma'}$, $\eta_1 = (1, 0.7954, 0.5907)$, $K_2 = (1, 1.5036, 0.5036)_{\gamma'}$, $\eta_2 = (111)_{\gamma'}$, $s = 0.26$.

2. In spite of the ordered structure, the original structure can be restored by the above twinning mode and simple and small shuffles, thus providing a theoretical background for experimental evidences such as the mobile nature of twin boundaries and the absence of extra spots in electron diffraction patterns.

3. {101}$\gamma'$ twins are considered to be variants of martensite rather than a lattice invariant strain in a variant of martensite.

4. In spite of (3), {101}$\gamma'$ twinning can be treated by the theory of deformation twinning, and no structural change is introduced by the twinning with the required twinning shear and simple and small shuffles. This is consistent with the experimental findings.

5. No distinction can be made between the mechanism of deformation twinning and that of transformation twinning in the present martensite, although the structure is ordered.

6. It is suggested that the absence of {1012} twinning as a lattice invariant shear in most hexagonal martensites is due to the fact that no solutions of the phenomenological theory exist for $\delta$ nearly equal to unity.

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I. Introduction

Twinning is closely related to the martensitic transformation, since most martensites are internally twinned as a result of lattice invariant shear and two variants of some martensites are twin-related in order to accommodate the constraints from the surrounding matrix (1). Thus the investigation of twinning mechanism is important for the clarification of the transformation mechanism and the understanding of the mechanical behaviors such as the shape memory effect (2)–(4) associated with the martensitic transformation. The twinning modes in $\gamma'$ Cu–Al–Ni martensite were investigated in previous papers (5)–(9), and two twinning modes, {121}$\gamma'$ and {101}$\gamma'$, twinnings, were found, but detailed analysis of the twinning mechanism was not given. As explained in the following, the twinning mechanism in the present martensite is of interest from a general point of view also, since the martensite is ordered and its sublattice is HCP. Laves pointed out that the B2 type lattice changes into an orthorhombic one by twinning if the same twinning mechanism is applied as that in the disordered BCC lattice (6), and suggested that the twinning in ordered lattices would be difficult. Later Cahn and Coll confirmed that Fe$_3$Al alloy with the DO$_3$ type structure shows resistance to twinning if the degree of order is increased, and that the alloy does not exhibit twinning if it is fully ordered (7). However, the present martensite is twinned and the twin boundary is mobile (8), although it is ordered. Does this fact mean that the twinning mechanism in the present martensite is compatible with the ordinary mechanism of deformation twinning though it is ordered, or that a different mechanism of twinning is necessary for transformation twins (8)–(9)? In HCP metals a number of twinning modes are predicted and many of them are experimentally found in deformation twinning (10). Among these, {1012} twinning is most fundamental and is found in all metals and alloys except for Cu–Ge alloy (11)–(12), since it is associated with a small twinning shear and a simple shuffle mechanism. Whereas the mode established as transformation twinning in martensites is only the {1011} mode, which is rather rare as a deformation twinning mode except in Cu–Ge alloy, although the presence of {1012} twins are confirmed in the present martensite (5). This difference is noteworthy, and the reason for it is to be pursued.

Keeping these general problems in mind, the present paper treats the following problems in $\gamma'$ Cu–Al–Ni martensite from crystallographic aspects based on the phenomenological theory of martensitic transformations (13)–(17) and the theory of deformation twinning developed by Bilby and Crocker (18).

1. Determination of twinning elements $K_2$, $\eta_1$, $\eta_2$ other than $K_1$.
2. Nature of {1012} twins. That is, to clarify whether the twins are really transformation twins or not.
3. Effect of order on twinning or related shuffle mechanisms.

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† In the present paper, the term transformation twins are defined as those introduced in martensites as a lattice invariant shear required in the phenomenological theory.
II. Summary of Experimental Investigations

The crystal structure of γ' (Cu, Ni)₃Al martensite is of the Cu₃Ti type with an orthorhombic unit cell, the lattice parameters being \( a = 4.382 \, \text{Å} \), \( b = 5.356 \, \text{Å} \), \( c = 4.222 \, \text{Å} \). So the structure reduces to HCP if ordering is disregarded, although small deviations from the HCP structure exist due to the anisotropy of the ordered structure. Thus, in the following discussion, both axes, orthorhombic and hexagonal, are used for describing planes and directions in the γ' martensite. To avoid confusion, the Miller notation (3 indices) is used when referring to orthorhombic axes, while the Miller-Bravais notation (4 indices) is used throughout when referring to hexagonal axes. The conversion of twinning plane indices from hexagonal to orthorhombic in the γ' phase, and that from γ' martensite to β₁ matrix due to lattice correspondence are shown in Tables 1 and 2, respectively, for the use in the following descriptions.

The experimental observations concerning twins in the γ' martensite in the previous and later investigations are summarized as follows.

1. The two twinning modes, \( \{121\}_γ' \) and \( \{101\}_γ' \), were observed; the former was predominant and the latter was seldom observed. Both twinning modes appear either independently or concurrently.

2. The above \( \{121\}_γ' \) and \( \{101\}_γ' \) twinning planes correspond to \( \{1011\} \) and \( \{1012\} \) twinning planes in hexagonal axes, respectively. As seen from Table 1, \( \{201\}_γ' \) and \( \{122\}_γ' \) twinning planes also correspond to \( \{1011\} \) and \( \{1012\} \) twinning planes, respectively. However, a careful analysis by electron diffraction patterns and the trace analysis of the corresponding electron micrographs did not detect the presence of these twinning modes. This fact means that only the twinning modes whose twinning planes correspond to the mirror planes in the matrix phase are realized as transformation twinning (see Table 2), and that the two twin-related regions can also be recognized as variants of martensites to each other.

3. Twin widths vary from one martensite to another, and \( \{121\}_γ' \) twins are usually much thinner than \( \{101\}_γ' \) twins. The twin widths of the former are usually less than \( 0.5 \, \mu \), while those of the latter are usually \( 1 \sim 10 \, \mu \).

4. Electron diffraction patterns from two twin-related regions showed no extra reflections, indicating that no structural change is introduced by twinning upon transformation although the martensite is ordered.

5. Twins are mobile under stress, irrespective of whether they are introduced on transformation or later by stress.

III. Analysis of Twinning in γ' Martensite

1. \( \{121\}_γ' \) twinning

   (I) Twinning elements

   The presence of \( \{121\}_γ' \) twins in the γ' martensite is well established, but the twinning elements other than \( K_1 \) were not determined in the previous papers. According to the phenomenological theory which gives a satisfactory account of the crystallography of the martensitic transformation in the present alloy, the direction and magnitude of the twinning shear is uniquely determined if a twinning plane is specified. Those determined for the \( \{121\}_γ' \) twinning plane are shown in Table 3. On the other hand, the theory of deformation twinning by Bilby and Crocker predicts two twinning modes for \( \{101\} \) twins, one being with a rational \( \eta_1 \) and the other with an irrational \( \eta_1 \). Among these, the direction and magnitude of the irrational \( \eta_1 \) mode coincides with those determined by the phenomenological theory in the above. Thus, the twinning elements of \( \{121\}_γ' \) twins are uniquely determined as shown in Table 3. It is interesting to note that \( \eta_1 \) in the transformation twinning is irrational, although it seems well established experimentally and theoretically that \( \eta_1 \) in the deformation twinning is rational.

† The magnitude of twinning shear reported in Ref. (9) is different from the value in Table 3, and is to be corrected as shown here, since the rational \( \eta_1 \) was assumed in the previous calculation.

Table 1

<table>
<thead>
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<td>{122}_γ'</td>
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Table 2

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<td>{301}_β₁</td>
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<td>{322}_β₁</td>
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Table 3

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<th>( K_2 )</th>
<th>( \eta_2 )</th>
<th>( s )</th>
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(2) Shuffle mechanism

Since the structure of the γ’ martensite is ordered, the shuffle mechanism upon twinning was carefully examined, in order to see whether or not the twinning violates the original γ’ structure. Since HCP as a sublattice is a double lattice structure, two atoms of the same kind are grouped as a pair, and the centers of gravity of paired atoms are chosen as lattice points. Then the twinning shear determined in the above acts on the lattice points and the necessary shuffles are determined so as to restore the original structure.

For analyzing twinning mechanism, crystals are usually cut by the plane of shear perpendicular to $K_1$ and $K_2$ planes, but, in the present case, the cut by $K_1$ plane is more convenient because of $\eta_1$ being irrational. Figure 1 shows a motif unit in the planes parallel to the $(12\bar{1})_\gamma$ twinning plane. In the figure two symbols connected by short lines represent paired atoms, the larger ones lying 0.335 Å above the plane and the smaller ones lying below the plane by the same amount (compare the distance with the scale in the figure). Now, these $(12\bar{1})_\gamma$ planes are divided into two groups, one denoted by 1, 2, 3, ..., being below the twinning plane and the other one denoted by 1, 2, 3, ..., being above the plane, and the twinning plane separating the two regions are taken to be at zero. The points denoted by 1, 2, 3, 4, ..., 0 are projected points on $[0\bar{1}0]_\gamma$ axis and fix the origin of the motif unit in each layer. Thus, the $(12\bar{1})_\gamma$ planes are stacked in such a manner that the point $A$ in the motif unit assumes the positions labelled with even numbers, while the point $B$ assumes the positions labelled with odd numbers.

In the following discussion, the atoms below the twinning plane are fixed, and those above the twinning plane are sheared by the amount determined in the preceding section. Then the positions of sheared atoms in $n$ layer are compared with those of fixed atoms in $\bar{n}$ layer, and necessary shuffles are sought so that the atoms in the $n$ layer get into the positions of the mirror images of atoms in $\bar{n}$ layer. Figure 2(a) shows the atoms in $\bar{1}$ layer (closed symbols) and those in sheared position in 1 layer (open symbols), and the arrows are necessary shuffles. It is to be noted that the arrows lie in the twinning plane and they do not contain components normal to the twinning plane. Thus, the shuffles are very simple and small, and all atoms in layer 1 are brought into the positions of mirror images of the same kind of atoms in $\bar{1}$ layer. The shuffle mechanism of atoms in other layers with odd numbers are the same. The shuffle mechanism in 2 layer is shown in Fig. 2(b). In this case the shuffles have components normal to the twinning plane only. Again the shuffles are very simple and small, and those in other layers with even numbers are the same as this. Thus, $(12\bar{1})_\gamma$ twinning in γ martensite is fully explained by the theory of deformation twinning without violating the original γ structure, even though the ordered structure is taken into consideration. In spite of the Laves’ prediction, $(12\bar{1})_\gamma$ twinning in the γ martensite is possible. This is a theoretical support for the direct observation of the mobile $(12\bar{1})_\gamma$ twin boundaries and for the absence of extra reflections in electron diffraction patterns described in the previous sections.

Similar analysis can be applied to $(20\bar{1})_\gamma$ twinning.

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**Fig. 1** Motif unit in $(12\bar{1})_\gamma$ plane. Atoms represented by larger symbols lie 0.335 Å above the plane defined by lattice points at midpoints of paired atoms, and those represented by smaller symbols lie below the plane by the same amount. The points labelled by numbers, ..., 2, 1, 0, 1, 2, 3, ..., define the origin of the motif unit in the corresponding layers. These $(12\bar{1})_\gamma$ planes are stacked in such a manner that the point $A$ in the motif unit assumes the points labelled by even numbers and the points $B$ assumes the points labelled by odd numbers.

**Fig. 2** Shuffling mechanism in $(12\bar{1})_\gamma$ twinning. (a) Shuffles in 1 layer. Closed symbols represent atoms in the fixed 1 layer. Open symbols represent sheared atoms in 1 layer, and arrows show necessary shuffles. (b) Shuffles in 2 layer. Definition of symbols are similar to those in (a). Thus, the necessary shuffles are normal to the $(12\bar{1})_\gamma$ plane.
which is equivalent to \{121\}_γ twinning if ordering is disregarded. The result shows that the original structure cannot be restored by twinning if a similar shuffle mechanism to the above is applied, and that much larger shuffles are necessary to restore the original structure. Thus, the \{201\}_γ twinning is not expected to be active in the present martensite, consistent with the observations described in the previous section\(^1\).

2. \{101\}_γ twinning

1) Is \{101\}_γ twinning a lattice invariant shear?

The presence of \{101\}_γ twinning is well established, but its occurrence is quite rare. Thus, the calculation of the phenomenological theory was carried out by assuming \{101\}_γ twinning as a lattice invariant shear, in order to see whether or not the theory is consistent with the observed crystallographic data. The result is shown in Fig. 3. The interesting feature of the result is that solutions exist only for \(\delta \approx 0.977\) and 0.960, and that they do not exist for \(\delta = 1\). The allowable maximum value of \(\delta\) is 0.977, which means that the strain at the habit plane is as large as 2.3%. Besides the predicted habit planes for allowable \(\delta\), do not coincide with the observed ones, \{331\}_γ\(^2\). These considerations indicate that it is inadequate to take \{101\}_γ twinning as a lattice invariant shear. A possible interpretation will be that the \{101\}_γ twins are individual variants of martensite themselves rather than lattice invariant strain in a single variant of martensite. This view is consistent with the above-described observation that \{101\}_γ twins are as thick as a few microns.

It may be interesting to recall here the general problem described in Sec. I. It was stated that \{1012\}_γ twinning is not likely to be a lattice invariant shear although it is a fundamental mode in deformation twinning. This fact can be explained by the result of the above calculations that no solution exists for \(\delta\) nearly equal to 1. Although the range of \(\delta\), for which solutions exist, vary with lattice parameters of the matrix and the martensite of the specimens concerned, the above result may be correct as a tendency in most BCC to HCP transformations. Therefore, it may be suggested that the reason for the absence of \{1012\}_γ twinning as a lattice invariant shear in most hexagonal martensites is simply because of the large elastic energies required at the matrix-martensite interface when it is introduced as such.

2) Shuffle mechanism

Even though \{101\}_γ twins are variants of martensite to each other, the movement of their boundaries may follow the theory of deformation twinning once they form. Thus, the theory is applied to the \{101\}_γ twinning in the present martensite. The twinning element of this mode is well known, and is shown in Table 4. Again the shuffle mechanism was examined by cutting the lattice by \(K_1\) plane. In this case \{101\}_γ planes consist of two kinds of planes, one consisting of the same kind of atoms (layers labelled with odd

\(^1\) Wasilewski\(^9\) suggested that the mechanism of transformation twinning might be different from that of deformation twinning on the ground of Laves’ prediction described in Sec. I. However, no distinction can be made between the two mechanisms in the present martensite, since the \{121\}_γ twinning in the ordered \(γ\) martensite can be explained both by the theory of deformation twinning and by the equivalence of lattice correspondences in the two twinned regions (II. (2)).
The analysis of \{122\}_γ twinning, which is equivalent to \{101\}_γ twinning in disordered state, by the theory shows that the original \γ structure cannot be restored if a similar shuffle mechanism is applied, and that much larger shuffles are required in order not to destroy the original structure. Thus, the presence of \{101\}_γ twinning and the absence of \{122\}_γ twinning can be explained by the effect of order on deformation twinning, as well as by the view that \{101\}_γ twins are variants of martensite to each other. Therefore, no distinction can be made between the two mechanisms at present.

Acknowledgment

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REFERENCES

(6) F. Laves: Naturwissenschaften, 39 (1952), 546.

Table 4

<table>
<thead>
<tr>
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<th>(K_1)</th>
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Fig. 4 Shuffling mechanism in (101)\γ twinning. Symbols are similar to those used in Figs. 1 and 2. (a) Shuffles (\(\uparrow\)) in 1 layer. (b) Shuffles in 2 layer are normal to the (101)\γ plane.

numbers) and the other consisting of a mixture of different kinds of atoms (layers labelled with even numbers). Again two of a paired atoms are situated 0.2533 Å above and below the (101)_γ planes. The atom positions in \(n\) layer and the sheared positions of atoms in \(n\) layer are shown in Fig. 4(a) and (b) for two kinds of layers, respectively. The necessary shuffles in Fig. 4(a) are indicated by arrows, and those in Fig. 4(b) are normal to the (101)_γ plane. It is seen from this figure that the shuffles are very simple and small, and that no structural change occurs by twinning. Further, the shuffle mechanism is the same as that considered to be acting in the deformation twinning of a disordered HCP structure\(^{24}\). Thus, the direct observation of mobile (101)_γ twin boundaries has been rationalized by the theory of deformation twinning.

\(\{122\}_\gamma\)