Analysis of the Transformation Strain of Bainites in Cu–Zn–Al Alloys by Utilizing Interference Fringes in TEM Images^{*1}

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In order to clarify the mechanism of the bainitic transformation, a new technique utilizing interference fringes in transmission electron micrographs was applied for observations of bainitic precipitates in copper alloys. Based on the theoretical consideration of image formation, the appearance of these fringes along the interface of a precipitate was taken to be evidence of the existence of a shear-type transformation strain. From the analysis on the spacing of interference fringes, the direction and magnitude of the shear strain were obtained. It was found that the shear strain associated with bainites has almost the same crystallographic direction and magnitude as the one associated with martensites in the same alloy. On the basis of these findings, a new model on the transformation mechanism is presented, in which the dislocation motion bringing about the lattice transformation is controlled by the atomic diffusion through a local concentration change. This model consistently explains both of existence of the transformation strain and the diffusion-control nature of the transformation.

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1. Introductionn

The study on the mechanism of the bainitic transformation has a long history, in which there has been a lot of controversy on the role of lattice shearing in the transformation process.¹⁻⁶⁾ In previous papers,⁷⁻⁹⁾ it has been shown that the bainitic transformation in a copper alloy is significantly accelerated by the application of an external stress. This result strongly suggests that transformation products are associated with a transformation strain, probably of the shear type. The applied stress must do work through this transformation strain and prompt the transformation. In order to clarify the nature of the transformation strain more precisely, detailed observations by the transmission electron microscopy has been planned in the present work. Especially we make use of an experimental technique developed recently,¹⁰⁾ in which a special kind of interference fringes are utilized. This technique should reveal shear strain associated with plate-shaped precipitates and enable one to measure the strain quantitatively. We will apply this technique for small bainitic plates produced at the early stage of the transformation.

There are several previous works which have suggested the existence of the transformation strain in bainites by electron microscopic observations. In a work,¹¹⁾ a stress field due to a bainitic plate was deduced by observing the shape of intersecting plates. In another work,¹²⁾ a strain field associated with a bainitic plate was inferred from observations on some irregularities in extinction contour lines. Preliminary observations on interference fringes along interfaces of bainites have been reported by the present authors.¹³⁾ In the present work, more detailed observations and elaborated analyses are presented. The results provide direct evidence on the existence of a shear-type transformation strain. In the

latter half of the present paper, on the basis of the present observations, we propose a new model on the transformation mechanism, which is able to explain both of existence of the shear strain and the diffusion-control nature of the transformation. The latter nature has been deduced from the composition analysis of transformation products by means of the analytical electron microscopy.^{14–20)} This nature has also been suggested in a previous paper,⁸⁾ in which the activation energy associated with the bainitic transformation has been measured. Traditionally, the problem setting on the transformation mechanism of bainites has been such an alternative one to ask which is the true mechanism between the shear process or the diffusional process. In contrast, the model shown later takes both processes to be indispensable in a single mechanism. Previous models will be discussed in the light of this new view in the last part of the paper.

2. Experimental Procedure and Results

Rolled sheets of Cu–28.9 at%Zn–6.1 at%Al were heattreated at 1123 K for 300 s and then quenched into water. This procedure brought specimens into a homogeneous β phase (of the ordered bcc structure). In order to induce the bainitic transformation, specimen sheets were aged in an oil bath at 433 K for 60–100 ks. By this procedure, the transformation was expected to proceed to 5–10% of the saturation level.⁸⁾ Disc specimens cut out from these sheets were thinned electrolytically in a solution of phosphoric acid saturated with chromic acid. They were observed by an electron microscope (JEM200CX) at the accelerating voltage of 200 kV.

Since the aging period was rather short, observed bainitic precipitates were all in an early stage of growth. It is well known²¹⁾ that the crystallographic structure of bainitic precipitates in copper alloys is the 9R type, which is the same as that of martensites in the same alloys. This was confirmed in the present experiments also. More precisely, it was identified as the disordered 9R structure, while marten-

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sites has the ordered 9R structure. In general, bainitic precipitates have a shape of thin plate, whose habit plane is approximately parallel to {12 11 2} of the matrix bcc crystal. (Hereafter, all crystallographic indices are given in the axes of the matrix crystal). This form of habit planes is also the same as that of martensites. Figure 1(a) shows a typical example of paired plates. Two plates are connected at a line to make a hinge shape having an obtuse angle. In this figure, each plate takes a rectangular shape which was cut out from an original broad plate by the upper and lower surfaces of the specimen. Such paired plates of bainites in copper alloys have been reported in many previous papers.⁷⁾ In contrast, such hinge-shape pairs of plates are hardly observed in the martensitic transformation. It is known that a hinge-shape pair is composed of two bainitic variants whose habit planes have indices, $\{12 \ 11 \ 2\}$ and $\{12 \ 11 \ \overline{2}\}$, respectively. These variants are in a twin relation of which the mirror plane is parallel to $\{0 \ 0 \ 1\}$ plane. The junction line between the plates is approximately parallel to $\langle 1 \ \overline{1} \ 0 \rangle$ direction. In this figure, interference fringes appearing in bainitic plates may be obvious. They are roughly parallel to the edges of the plates and densely packed towards the edge on the left end. Since they have a nature of equal-thickness contours, as shown later, we can deduce lenticular shapes of these plates from observations of these fringes. These fringes appear only when a picture is taken in a Bragg reflection of the matrix crystal. They never appear in a picture taken in a reflection of

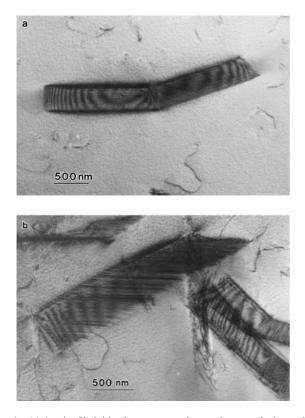


Fig. 1 (a) A pair of bainitic plates connected at an obtuse angle, imaged in a reflection of the matrix crystal, whose reflection indices are $g = 1 \bar{1} 0$. Many interference fringes can be seen, which have a nature of equal-thickness contours. (b) Bainitic plates with various shapes. In a plate at the center, a reflection of the bainite is excited, in which many stacking faults, appearing as dense parallel lines, can be seen in addition to interference fringes.

bainites. For these plates, crystallographic analyses were performed by means of electron diffraction. We identified the habit planes of plates on the left and the right in Fig. 1(a) as $(12\bar{2}11)$ and (12211), respectively. The junction line of the plates is parallel to $\begin{bmatrix} 1 & 0 & \overline{1} \end{bmatrix}$ direction. Figure 1(b) shows another example of bainitic plates, in which, in a plate at the center, a reflection of the bainite is excited in addition to a reflection of the matrix. It can be seen that the plate contains many stacking faults which are parallel to the basal plane of the 9R structure. Stacking faults were almost always observed when a bainite was imaged in a reflection of the bainite itself. Note that the inner structure of dense faults is also a feature of martensites. Using an image of a plate in an upright position, we can measure the plate thickness. The measured thickness in the present specimens was in the range of 30-50 nm. Thus, bainitic precipitates were quite thin and had a large aspect ratio, roughly 100 or more. It is also noted that those plates have sharp edges. Measured wedge angles of edges were in the range of $2-3^{\circ}$. The thickness of the left plate of the pair shown in Fig. 1(a) was measured more precisely by taking a series of pictures near in an upright position. The thickness was determined as the smallest image width of the plate among those pictures. We have obtained the thickness 31 nm at the thickest part of the plate. This value will be used in an analysis in a later section.

Figure 2 shows a pair of bainite plates observed in various reflections. Interference fringes appear clearly. One plate seems to have a shape of semi-circle, but the plates are cut by the upper and lower surfaces of the specimen and terminated by irregular steps at one end. The morphology is sketched in Fig. 2(f). Two plates ABYX and CDYX (called Plate 1 and Plate 2, respectively) are connected at the junction line XY. The intersecting lines of the pair with the upper and lower surfaces are AXC and BYD, respectively. Firstly, crystallographic features of these plates were examined by use of diffraction patterns. The surface normal of the specimen in the observed area was roughly parallel to [0 0 1] direction of the bcc matrix. The habit planes of Plate 1 and Plate 2 were identified as $(2 \ 12 \ 11)$ and $(\overline{2} \ 12 \ 11)$, respectively. The normals of these habit planes are denoted by H_1 and H_2 hereafter. The junction line XY is approximately parallel to $\begin{bmatrix} 0 & 1 & \overline{1} \end{bmatrix}$ direction. As shown later, the shear direction (S) of these bainite plates is nearly parallel to this junction line. The crystallographic orientations of these bainites are shown in Fig. 3. In Fig. 2, it may be obvious that the fringe spacing is different for each reflection. In (a), the spacing in Plate 1 is closer than that in Plate 2. On the contrary, the spacing in Plate 2 is closer in (b). In (c), the fringe spacing is almost the same between two plates and seems to be closer than those in (a) and (b). In (d), the spacing in two plates is rather similar to that in (c). Fringes are hardly visible in (e). This is due to a special direction of the reflection vector used, which was almost perpendicular to the shear direction of the bainite. All these features of pictures can be explained by an equation given in a later section.

3. Analysis of Interference Fringes

In this section we show how interference fringes appearing in the above photographs are related to the intrinsic nature of

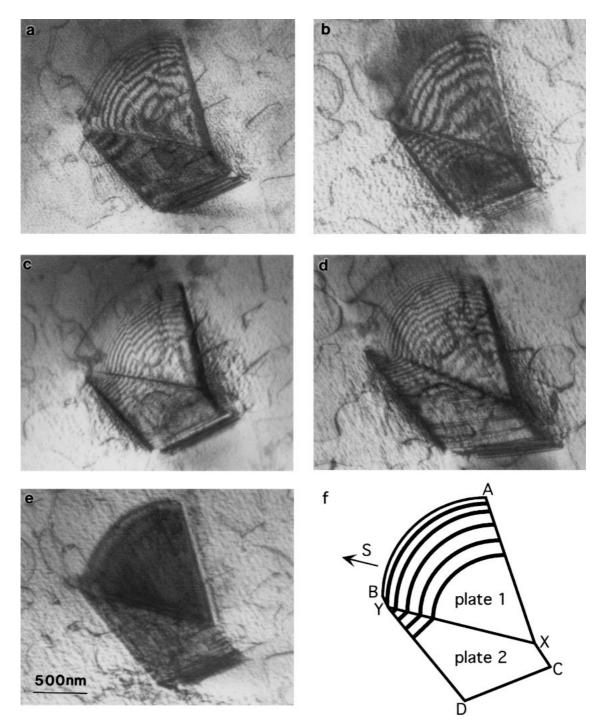


Fig. 2 Paired bainitic plates, observed in various reflections (g). Two plates, Plate 1 and Plate 2, are connected at the junction line XY. The approximate shear direction is denoted by S. (a) $g = 1 \ 1 \ 0$, (b) $g = \overline{1} \ 1 \ 0$, (c) $g = 0 \ 2 \ 0$, (d) $g = 0 \ 1 \ \overline{1}$, (e) $g = \overline{2} \ 0 \ 0$, and (f) a sketch of the plates.

the bainitic transformation. Evidence will be presented on the existence of a shear strain associated with a bainite plate. For this purpose, we first give the explanation on the mechanism how the fringes are produced. Previously, similar interference fringes were observed in the case of wedge-shaped martensites embedded in the matrix crystal.^{22,23)} Their origin has been attributed to the shear strain associated with the martensitic transformation. A detailed explanation of the fringe image has been given on the basis of the dynamical theory of the electron diffraction in a previous paper.¹⁰⁾ Here we only describe the essential point of the explanation.

Figure 4 shows the geometry of observations in the electron microscopy. A thin bainitic plate is inscribed in a foil specimen. The plate has a wedge shape, of which the local thickness measured perpendicularly to the habit plane is denoted by *t*. In the figure, the lower surface of the wedge is taken to be the habit plane. An important factor in the image formation of electron micrographs is the phase change of an electron wave during passing through a defective part, *i.e.*, the wedge plate in the present case. We assume that the matrix crystal is in an exact Bragg reflecting condition but that the wedge plate is completely out of the condition. Then

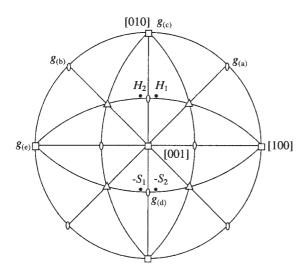


Fig. 3 Stereographic representation of crystallographic directions in observations of Fig. 2. The reflection vectors (g) used in the observations are shown. The habit plane normals for Plate 1 and 2, H_1 and H_2 , and the shear directions for these plates, S_1 and S_2 , are also shown.

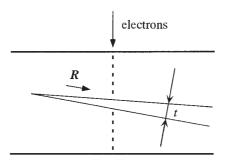


Fig. 4 Geometry of the TEM observation in the present experiment. A wedge-shaped baintic plate is inscribed in a foil specimen. The plate is homogeneously sheared, so that the upper surface of the plate is displaced with respect to the lower in the direction \mathbf{R} . The local thickness of the plate (*t*) is measured perpendicularly to the habit plane (the lower surface).

the phase change (α) is given by the expression,

$$\alpha = 2\pi g. \boldsymbol{R} \tag{1}$$

where g is the reflection vector and R is the displacement vector of the upper interface of the plate with respect to the lower interface. This equation may be recognized by an analogy to the well-known case of stacking faults. We further assume that the plate is associated with a shear-type transformation strain, as in the case of martensites. Now suppose that such a plate is newly produced in the matrix. To make the new lattice, the crystal in the plate should be deformed by shear with keeping the coherency at the interfaces. In this process, any points on the upper interface of the plate are displaced with respect to the lower. The lower interface (the habit plane) is assumed to be stationary. The shear direction lies in the habit plane. The displacement is obviously given by

$$\boldsymbol{R} = t\gamma \boldsymbol{S},\tag{2}$$

where *S* is a unit vector which is parallel to the shear direction and γ the magnitude of the shear strain. When the phase change of eq. (1) amounts to zero or integral multiples of 2π , no special contrast arises in the micrographic image.

When it is equal to π or odd multiples of π , then a dark fringe appears. This is the essence of the formation mechanism of the observed fringe system. Since the phase change as well as the displacement increases linearly with the local thickness, dark fringes appear periodically along the wedge plate. From these considerations, it can be easily concluded that a dark fringe appears at the point where the local thickness fulfills the condition,

$$t = (n - 1/2)t_0, (3)$$

where *n* is an integer (n = 1, 2, 3, ...) and the basic thickness period t_0 is given by

$$t_0 = 1/(\gamma |\boldsymbol{g}.\boldsymbol{S}|) \tag{4}$$

A more detailed deduction¹⁰ has shown that there is a small additional term in the denominator, which depends on the deviation from the exact Bragg condition. Since its magnitude is small in the case that the Bragg condition is well fulfilled, it will be neglected in the following. Also there is a small geometrical factor which is related to the plate thickness measurement and which arises from the fact that the upper and lower interfaces are not exactly parallel to each other. Since the effect of this factor is small for thin wedges, it will be neglected. From eq. (4), it may be recognized that the direction as well as the magnitude of the reflection vector affects the fringe spacing, which is proportional to t_0 . The fringe spacing becomes closer for a reflection whose vector is parallel to the shear direction. For example, the reflection vector in Fig. 2(d) is almost parallel to the shear direction. Therefore, the spacing is fairly close in the figure. The one in Fig. 2(c) is deviated by about 45° from the shear direction but has a large magnitude, so that the spacing in (c) is roughly equal to that in (d). Similarly, other features of Fig. 2 can be explained by eq. (4). The above explanation clearly shows that these fringes have the nature of an equal-thickness contour. Actually they can be used to measure the local thickness of a plate with a fairly good precision, since the value of t_0 is small, typically 1–3 nm, as shown later. They should be distinguished from another type of equal-thickness fringes. The latter fringes, whose period corresponds to the extinction distance of the electron wave, are commonly observed in a wedge-shaped specimen.

The above explanation indicates that the appearance of the interference fringe system is closely related to the existence of the shear strain in the bainitic plate. Only a homogeneous shear gives the linear dependence of the displacement on the local thickness and produces regularly spaced fringes. Therefore, the appearance of these fringes can be taken to be evidence of the lattice shearing during the transformation. Since in the present experiments such fringes have been always observed when a bainitic plate is imaged in a matrix reflection, we conclude that bainites are surely associated with the transformation shear. This is the most important conclusion of the present experiments. As mentioned above, observed fringe systems are quite similar to those observed in martensite plates. Therefore the lattice deformation in the bainitic transformation may be similar to that in the martensitic transformation. We next examine the shear strain more quantitatively.

In order to obtain the direction and magnitude of the shear

strain associated with a bainitic plate, we make use of eqs. (3) and (4). First we try to find the shear direction only. For this purpose, we do not need to measure the local thickness of a wedge plate. We need three independent reflection vectors to obtain three components of the vector S in eq. (4). Here we take Plate 1 in Fig. 2 and choose three photographs, Figs. 2(a), (b) and (d). In order to obtain relative values of t_0 , we counted the number of fringes lying between the peripheral point B and a fixed point which was chosen in the vicinity of the mid point between points X and Y. A caution was taken for spurious fringes appearing outside the plate, which were caused by a strong elastic strain around the plate tip. Obtained numbers were n = 8.5, 6.0 and 14.5 for reflections, $g = (1 \ 1 \ 0), (\overline{1} \ 1 \ 0)$ and $(0 \ 1 \ \overline{1})$, respectively. The reciprocal of (n-1/2) corresponds to a relative value of t_0 . Substituting these values in eq. (4) and solving the resultant simultaneous equations, we obtained the direction cosines of the shear vector as S = (0.125, 0.676, -0.726). This direction is shown in Fig. 3 (denoted by S_1). The shear direction of Plate 2 has been deduced by taking into account the twin relation between these plates. The direction is denoted by S_2 in Fig. 3. This direction was found to be consistent with the observed spacings of interference fringes in Fig. 2. As far as the present authors are aware, there is no report on the experimental measurement of the shear direction of bainites so far. We compare this result with the corresponding one for martensites in copper alloys having a similar composition. There are several results obtained experimentally²⁴⁾ or theoretically.²⁵⁾ A theoretical calculation is performed by use of the so-called phenomenological theory of martensitic transformation. These results are in a fairly good agreement with each other. Here we take the theoretical value in Cu-31.6 at%Zn-4.5 at%Al alloy, which is S = (0.1410, 0.6485, -0.7481)for the same variant as Plate 1. It is to be noted that calculations by the phenomenological theory employ the data of lattice parameters, so that calculated results should be the same for those alloys having the same lattice parameters. In the present case, differences in the lattice parameters between two Cu-Zn-Al alloys used are negligible. The deviation of the shear direction (S_1) for bainites from this direction for martensites is about 2.1° in arc. Therefore it is concluded that the shear direction of bainites agrees quite well with the one of martensites.

Next we try to find the magnitude of the shear strain. For this analysis, we make use of Fig. 1(a), in which the thickness of the bainitic plate was measured to be 31 nm at the thickest part, as mentioned in a previous section. We have judged that the number of fringes between the plate tip and the thickest point is about 14.5. Therefore we obtained $t_0 = 2.21$ nm from eq. (3). We use the shear direction obtained above, which corresponds to S = (0.676, -0.125, -0.726) for the bainite variant in hand. We also need the lattice parameter of the matrix crystal, which has been reported²⁵⁾ to be a = 0.294 nm. Substituting these values and the reflection vector $\mathbf{g} = (1/a)[1, -1, 0]$ into eq. (4), we obtained $\gamma = 0.166$. Again we compare this value with the corresponding one of martensites in a Cu-Zn-Al alloy. The theoretical value of the shear strain is $\gamma = 0.198$ in Cu– 31.6 at%Zn–4.5 at%Al alloy.²⁵⁾ Comparing these values, it may be judged that the magnitude of the shear strain obtained for bainites is in a reasonable agreement with the corresponding value for martensites, if we take into account the approximate nature of the present analysis. Thus, in copper alloys, bainites have similarity with martensites on many aspects, such as the crystallographic structure, the plate shape with {12 11 2} habit plane, the internal defective structure and in addition the associated transformation strain.

4. An Atomic Model of the Transformation Mechanism

As shown in the above, experimental evidence has been obtained on the conclusion that bainitic precipitates are associated with a transformation strain which is of the shear type. On the other hand, there is plenty of evidence for the fact that the rate of the bainitic transformation is controlled by the diffusion of solute atoms.^{8,14–20} Now, we discuss how these two contributions, shear and diffusion, are incorporated in a single transformation mechanism. It will be shown that they are compatible at the atomic level. A rough sketch of the present model was previously presented by one of the present authors.²⁶ Here the model will be given in a more detailed form.

Since any plastic shear deformation can be produced by movements of dislocations, a bainitic precipitate should be represented by an array of dislocations, which is schematically shown in Fig. 5. A precipitate in the matrix is depicted as an ellipsoid, whose broad face is parallel to the habit plane of the precipitate, though actual bainitic plates have a much flatter shape. As depicted by arrows in the figure, the invariant plane of the associated shear is parallel to the habit plane. To visualize dislocations, the ellipsoid is sliced by layers parallel to the habit plane. Each layer is bounded by a dislocation loop, of which the Burgers vector points to the shear direction. The motion of these dislocations (so-called transformation dislocations) makes a new lattice by shearing the matrix lattice. These dislocation should be a kind of partial dislocations, since the motion of a complete dislocation does not produce a new structure but leaves the lattice unchanged. The magnitude of the Burgers vector is given by

$$b = \gamma d, \tag{5}$$

where γ is the magnitude of the shear strain and *d* the layer thickness. Although the habit plane and the direction and magnitude of the shear strain are unique for the bainitic

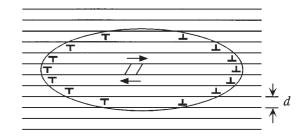


Fig. 5 A schematic illustration of an ellipsoidal precipitate (bainite) created in the matrix. The lattice in the ellipsoid was transformed by shearing from the matrix lattice. This transformation can be modeled by an array of transformation dislocations. Layers having the thickness d are parallel to the habit plane of the bainite.

transformation in a particular alloy, there is an arbitrariness on the selection of the layer thickness. However, the selection does not affect the conclusion of the following arguments. It is also to be noted that the core structure of those dislocations is rather complicated, since the transformation shear is composed of a combination of the lattice transformation (socalled Bain distortion) and the lattice invariant shear in the case of bainites or martensites in copper alloys. Even the atomic arrangements in the habit plane, *i.e.* {12 11 2} plane, are not so simple. However, in the present analysis we only needs the transformation shear (so-called the macroscopic shear) as a whole and does not need to consider any details.

Now we examine how the motion of these dislocations is controlled by the diffusion of component atoms in the alloy. For simplicity, we take a binary alloy, in which the average concentration of solute atoms is x_0 . In the situation shown in Fig. 5, we assume that the equilibrium solute concentrations of α phase (bainite) and β phase (matrix) are x_{α} and x_{β} , respectively. In the case of copper alloys, the relation, $x_{\alpha} < x_0 < x_{\beta}$, holds. Therefore, it is necessary for the precipitate to grow, or for a transformation dislocation to move, that some solute atoms flow out from (or some solvent atoms flow into) the precipitate. Since solute atoms flown out from the precipitate stay in its vicinity for a while, the solute concentration just outside of the precipitate may be rather high, probably near to the equilibrium value (x_{β}) . On the other hand it has a lower value (x_0) in the distance. Therefore, a diffusional flow of solute atoms to the distance occurs naturally. This flow should control the growth rate, as in the genaral case of the precipitate growth.

In order to examine the situation more closely, a part of the ellipsoid is illustrated again in Fig. 6, in which the atomic arrangements around a transformation dislocation lying in an interface are schematically shown. On the left and the right of the dislocation, there are α phase and β phase, respectively, in the same layer. If each side of the dislocation has the respective equilibrium concentration of solute atoms, the dislocation should be stationary. If on the other hand there is any deviation from the equilibrium on one side, the dislocation would be attracted toward that side, since the

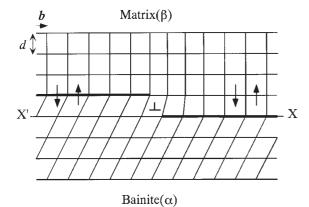


Fig. 6 An atomic view of the interface between a bainite and the matrix crystal. The interface (denoted by a thick line) has a step at the center of the figure, where a transformation dislocation exists, whose glide plane is parallel to XX' and whose Burgers vector is denoted by b. The vertical arrows represent diffusional atomic flows.

deviation always gives rise to an excess free energy. For example, suppose that the dislocation moves suddenly to the left in the figure without any atomic diffusion. Then it creates an area of β phase having a concentration lower than the equilibrium (*i.e.*, x_{α} instead of x_{β}). This area should exert a force on the dislocation to pull back to the original position. This situation is rather similar to the surface tension associated with a stacking fault, of which the excess energy per unit area corresponds to the force on the unit length of a peripheral dislocation. In the actual situation, there are always atomic flows of solute atoms from the vicinity of the dislocation to the distance, as mentioned above. Therefore, a transformation dislocation perceives a force from the adjacent β phase region, where the solute atom concentration is incessantly changing. The α phase region also can exert a force on the dislocation if there are any concentration changes. These situations can be summarized in the following expression, where the resultant force f (to the right) on the unit length of the dislocation is given. That is,

$$f = d(\Delta G_{\beta} - \Delta G_{\alpha}), \tag{6}$$

where ΔG_{α} and ΔG_{β} are the excess free energies per unit volume of the layer on the left and on the right, respectively. This expression has been deduced rather intuitively by the analogy to the case of stacking faults. Maybe, a more detailed discussion based on the thermodynamics is necessary for rigorous deduction of this force. There may be some difficulties for the actual evaluation of eq. (6) because of the non-linear relation between the concentration and the free energy. Also there remains another problem to choose the width of the area in which the concentration is evaluated. For the actual dislocation motion, we must take into consideration another force, a frictional force due to the basic nature of the crystal lattice, which corresponds to Peierls force for an ordinary dislocation. The evaluation of this force may be quite difficult because it is related to two lattices. Even though there are these difficulties in evaluating the actual force, it may be recognized that this equation has a feature that can explain the expected control action of the atomic diffusion on the dislocation motion. This is the essence of the present model on the atomic mechanism of the diffusioncontrolled dislocation motion.

We further examine the above model more closely in several points. First we note the fact that all the lattice points are conserved in the course of a dislocation motion (we here consider a glide motion only). Obviously, any dislocation motions never create nor eliminate a lattice point. Therefore, we are safe to say that the correspondence between the parent and transformed lattices exists on all lattice points, as far as we consider a dislocation process of transformation. Another noticeable point of the dislocation model is the interaction between dislocations. Since all dislocations have the same Burgers vector, as shown in eq. (5), they are repulsive with each other. Therefore, an ellipsoid having transformation dislocations is necessarily flattened to a thin plate with sharp edges. This feature is in accord with present observations of bainitic precipitates. It is to be noted that the plate shape itself exerts influence on its growth rate, since the growth rate is inversely proportional to the radius of curvature of the plate tip.²⁷⁾ In copper alloys, there are other kinds of precipitates which are produced at a higher temperature. They have a blocky shape instead of a plate.²⁸⁾ This shape suggests that they are created not by a dislocation mechanism but by a different one. One reason for the fact that bainites becomes predominant over other precipitates at relatively low temperatures should be attributed to its high growth rate.

Next we look at lattice sites responsible for the atomic diffusion. Taking into account the randomness of the atomic migration, it may be evident that any lattice points in the interface between two phases can be possible sites for flow-in or flow-out of solute atoms. Vertical arrows in Fig. 6 indicates these flows. Thus, the concentration of solute atoms in a precipitate is to be determined by an average of atomic flows over a fairly broad area. On the other hand, atomic movements associated with the creation of a new lattice (i.e. shearing movements) occur exclusively at the dislocation core. Therefore, atomic movements to bring about the composition change are obviously independent from those associated with the lattice transformation. In other words, two processes, the atomic diffusion and the lattice shearing, are compatible with each other by nature. This view does not seem to be fully accepted in several previous papers, as shown in a later section.

The present model does not contain the thickening process of plates. For this process, the nucleation of a dislocation loop on a broad interface of a precipitate is necessary. The nucleation may require a greater deviation from the equilibrium concentration in the vicinity of the interface in comparison with the case of dislocation motion. We need more elaborated discussions on this process, which are postponed to a later publication.

5. Discussion: Comparison with Other Models

There has been a long standing debate on the mechanism of the bainitic transformation, as described already. The manner of weighing the two mechanisms, lattice shearing process and diffusional reconstruction process, to the transformation was the main subject of the debate.^{3–6)} As shown in the previous section, the view of the present paper is such that these mechanisms are compatible with each other and they are combined intimately in a single transformation mechanism. This is in a sharp contrast with views in the past. Therefore, from the present view point, most of previous models are somewhat improbable. In the following, we show how the present model is different from previous ones. Discussions will be confined mainly to the transformation mechanism in copper alloys.

The first systematic study on the growth of plate precipitates (bainites) in copper alloys has been done by Garwood.^{29,30)} Based upon observations of crystallographic similarities between them and martensites in Cu–Zn alloy, he recognized the important role of shear in the growth. By noting the slow growth rate of bainites in comparison with martensites, he also concluded that the growth process is associated with the atomic diffusion. By that time Ko and Cottrell¹⁾ had presented a model of the transformation mechanism for bainites in ferrous alloys, in which the lattice shear to make the ferrite lattice from the austenite was assumed to be controlled by the diffusion of interstitial

carbon atoms. Garwood did not follow this model, but presented a two-stage model, because there was an influential view by Christian²⁾ that shear and diffusion processes may not be compatible with each other in substitutional alloys. It has been argued that in substitutional alloys a diffusive flow of atoms could destroy the correspondence between the parent and product lattices. Taking into account this incompatibility, in the two-stage model, it has been assumed that at the first stage the martensitic shear makes the new lattice and at the subsequent stage the diffusion of substitutional atoms brings about a composition change. In actual happening, the outer rim of a plate expands by a shear process and then the diffusional composition change follows in the bulk. This model is in contrast with the present model, in which the shear process as represented by a dislocation motion is taken to be compatible with the diffusion process even in substitutional alloys.

Several later papers seems to have been influenced by this two-stage model. There are variations of the two-stage model, $^{21,31-34)}$ in which the shear stage is separated from the diffusion stage in a time sequence. These models now seem to be somewhat inadequate in the light of recent experimental results. It has been shown by the analytical electron microscopy that a composition change to the equilibrium value is attained from the earliest stage of the transformation in noble metal alloys.14-20) This indicates that the bainitic transformation is a simultaneous process of the structural change and the compositional change. There is another type of model in which for ferrous alloys the shear and the diffusion is coupled in a single mechanism.^{35,36)} In the model, however, it was assumed that the concentration change is suppressed to some extent because of a rapid transformation. This assumption is obviously inadequate for the case of bainites in noble metal alloys. There are some other suggestions in which the coupling of the shear and the diffusion from the earliest stage of the transformation is emphasized.³⁷⁻³⁹⁾ However, the detailed atomic process has not been presented yet.

Although the models mentioned above take the shear process as the lattice transformation mechanism, there have been a few proposals which do not accept the shear process. Aaronson and coworkers have published several review articles on the plate-precipitate (bainite) formation.^{4–6)} In one of them,⁴⁾ two growth mechanisms have been defined as follows: "At atomic level, diffusional growth is described as individual, poorly coordinated, thermally activated jumps occurring in the manner of biased random walk, whereas growth by shear is taken to be tightly coordinated glide of atoms to sites in the product phase which are predestined to within the radius of a shuffle". Thus, in this definition, these two mechanisms were supposed to be incompatible with each other. Such an alternative view is quite improbable from the present view point, as mentioned already. They further discussed requirements for distinguishing the shear mechanism from the diffusional one in substitutional alloys. The requirements included "the absence of a composition change during growth". Applying this requirement to the case of plate precipitates in Cu-Zn alloy, they denied the shear process as the transformation mechanism.⁵⁾ This reasoning is not legitimate from the view point of the present paper, because the compositional change can be compatible with the shear process in the present model. In several papers, $^{4-6,40-43)}$ ledges in interfaces between a

In several papers,^{4–0,40–45} ledges in interfaces between a precipitate and the matrix are invoked as the growth front of the precipitate. They have been taken as sites where both the structure change and the composition change occur. This diffusion-controlled ledge mechanism has been often contrasted with the shear mechanism. However, descriptions have not been quite clear on the question whether ledges are associated with a stress field or not. In the ledge mechanism, it is commonly supposed that atomic sites responsible for the composition change are restricted only to sites in ledges. This supposition is rather questionable, as is described below.

Recently, Howe⁴⁴⁾ presented a new version of the ledge mechanism for the case of fcc/hcp transformation in Al-Ag alloy. In this model, a Shockley partial dislocation is taken as the ledge. Since the motion of a dislocation necessarily brings about shear deformation, this model is obviously different from other ledge models, in which lattice shearing is not admitted in the transformation process. At first sight this model is quite similar to the dislocation model presented in a previous section. However, there are several differences between these models. In this model, a partial dislocation is considered in a most simple case of hcp/fcc transformation. In the model presented in a previous section, transformation dislocations are assumed in a more general case, where the slip plane is not a simple lattice plane. More importantly, this model as well as other ledge models assumes that the composition change occurs only at sites in a ledge, or precisely those at a kink of a dislocation. According to this model, if an atom of an appropriate species happens to migrate to neighboring sites of a kink, then the kink moves and takes the atom into the precipitate. Thus the compositional change and the structural change occur strictly simultaneously. However, such a restriction on sites seems to impose a difficulty on the mechanism of composition adjustment of a precipitate. In this model, the adjusting mechanism is fully attributed to a narrow channel of a kink. That is, a kink would be supposed to have a special ability to judge whether a nearby atom of one species should be taken into the precipitate or not. Thus the site restriction seems to be too strict. This restriction may be related to the supposition that the precipitate is taken as a homogeneous continuum having a definite concentration. If we take into account the discrete nature of atomic arrangements instead of a continuum, we would necessarily confront local fluctuations of the concentration. For example, we cannot define the local composition for a single kink, nor for small number of atoms. In general, some local fluctuations should be always admitted in a thermodynamical situation and quantities such as the concentration can be defined only by averaging over numbers of atoms. In contrast to those ledge models, the model shown in the previous section seems more natural, in which the local concentration or the local free energy is defined by averaging over a wider area in the vicinity of a transformation dislocation and atomic flows are necessarily expected to occur at numeral sites in that area. Thus, the formulation given in the previous section seems to have a more plausible feature as the true mechanism.

6. Conclusions

- (1) A new technique utilizing interference fringes in transmission electron micrographs was applied to examine the nature of the transformation strain associated with bainitic plates in a Cu–Zn–Al alloy. The consideration on the origin of these fringes indicates that they are closely related to the shear-type transformation strain. Therefore, the appearance of these fringes was taken to be evidence of the existence of a shear strain associated with bainites.
- (2) From the analysis on the spacing of interference fringes, the direction and magnitude of the shear strain associated with bainites were deduced. We obtained the results that the unit vector of the shear direction is S = (0.125, 0.676, -0.726) and the magnitude of the shear strain is $\gamma = 0.166$. These values reasonably agree with those of the shear strain associated with martensites in the same alloy.
- (3) A new model on the mechanism of the bainitic transformation has been presented, in which the dislocation motion to bring about the lattice transformation is controlled by the atomic diffusion through a local concentration change, that is, a local change in the free energy. The new model consistently explains both of existence of the shear-type transformation strain and the diffusion-control nature of the transformation.
- (4) The new model was compared with various models presented so far. Most of previous models contains some improbable points because they are based on the inadequate supposition that the shear process and the diffusion process are incompatible with each other in the transformation mechanism.

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