Origins of Internal Structure in Massive Transformation Products

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The internal structure in massive phases formed during six massive transformations has been reviewed. A counterpart review has also been made for the proeutectoid ferrite reaction, mainly in alloy steels in which bulk partition of alloying elements between austenite and ferrite has not occurred. Both dislocations and twins comprise this structure unless the stacking fault energy is too high to permit twin formation. Volume and shape changes associated with transformation can explain dislocation loops through stress-induced displacement and multiplication of misfit dislocations into the softer phase by means of either a dissociation reaction followed by Ashby-Johnson prismatic looping or emanation of glide loops from Frank-Reed sources. Following Gleiter *et al.*, the "growth accidents" concept used to explain dislocation and twin formation during grain growth proves equally suitable for explaining formation of the same features during the massive and other diffusional transformations. Climb of interfaces produced by edge-to-edge rather than the usual plane-to-plane matching, introduced by Kelly and Zhang and experimentally supported by Nie and Muddle and by Howe *et al.* for the $\alpha \rightarrow \gamma_m$ transformation in near-TiAl alloys, is proposed as another source of dislocations in the product phase.

I. INTRODUCTION

A long-term problem in the mechanism of the massive transformation (MT) has been that of understanding the origin of the internal (*i.e.*, intragranular) structure, in the form of dislocations or twins, in the products of this transformation.[1] The papers published in this issue of *Metallurgical and Materials Transactions* as the proceedings of a symposium on "The Mechanisms of the Massive Transformation," held during the Fall 2000 TMS/ASM Meeting, have little to say about the origins of this structure. The authors accordingly decided to assemble an overview of both experimental observations on this internal structure and explanations advanced for its formation, and to offer their own contributions to these explanations. Concepts evolved from studies of internal structure produced during grain boundary migration and both experimental observations and explanations developed during counterpart studies on proeutectoid ferrite formation in steel, wherein the interstitial solute diffuses but substitutional solutes do not, will also be utilized. As a by-product of this effort, a suggestion will be made that the defects left behind by the massive transformation interface can in at least one case provide important clues to the interfacial structure and the operative boundary migration process.

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II. EXPERIMENTAL OBSERVATIONS ON INTERNAL STRUCTURE ASSOCIATED WITH THE MASSIVE TRANSFORMATION

Kittl and Massalski[2] studied the $\beta \rightarrow \zeta_m$ MT in Cu-Ga alloys with hot-stage optical microscopy and observed slip in the product phase during its growth. Baro *et al.*[3] noted that thin foils of Ag-24.5 at pct Al alloy underwent violent shaking during a hot-stage transmission electron microscopy (TEM) study of MT in this material. Dislocation emission from β:ζ_m interphase boundaries into the parent β phase in the Ag-Al system was observed by Baro *et al.*[3] and Baro and Perepezko.[4] The dislocation emission occurred when the undercooling below the congruent transformation temperature was greater than $\sim$50 K. This emission appeared to occur by the "peeling off" of a portion of the growth ledge into the β matrix, where it became a lattice dislocation. Growth ledges have been repeatedly observed in this transformation.[3,5]

Baro and Perepezko[4] found that at undercoolings in the range of 10 to 100 K, some dislocations at the β:ζ_m boundaries were left behind in MT product phase as the boundaries moved forward, a result consistent with the optical microscopy observations of Kittl and Massalski[2] on the $\beta \rightarrow \zeta_m$ MT in Cu-Ga alloys. These dislocations were often connected to the boundaries by stacking faults. The larger the undercooling, the higher the density of faults.

Delaey *et al.*[6] observed microtwins, large twins, and a high density of aligned dislocations in a Cu-37.6 at pct Zn alloy that had transformed massively from the β (bcc) to the α_m(fcc) phase. The complementary study of Malcolm and Purdy[7] showed that the β:α_m interface in a Cu-Zn alloy could generate dislocations under high driving forces, but that subsequent isothermal growth, now apparently in the form of α with a lower Zn concentration than in the matrix, resulted in a nearly dislocation-free product.

A number of studies have shown that the γ(fcc) $\rightarrow \alpha_m$(bcc) transformation in ferrous alloys produces dislocations in α_m.[8,9,10] The pioneering investigation of Hultgren and

Herrlander[11] on irons containing 0.004 to 0.05 pct C was probably made on ferrite formed as both proeutectoid ferrite and as massive ferrite. Careful optical microscopy disclosed the presence of "veining" (now recognized as sub-boundaries), particularly in rapidly cooled specimens (in which the transformation product is more likely to have been massive), as well as in specimens that had undergone externally imposed plastic deformation. Studies on the proeutectoid ferrite reaction in Fe-C-Mo alloys, in which carbon partitions between austenite and ferrite but Mo does not,[12,13,14] provide further relevant observations on this phenomenon. Marked reductions in the growth kinetics of proeutectoid ferrite as a result of the required long-range volume diffusion of carbon in austenite makes these kinetics markedly slower, thereby permitting microstructural evolution to be examined in more detail. The only substantive difference anticipated between the internal defect structures of ferrite formed massively and that developed as proeutectoid ferrite is that the slower growth kinetics during the latter mode probably provide additional opportunities for the density of transformation-induced defects to be reduced through various forms of annihilation and the reconfiguration of these defects into lower energy states. However, even this difference is mitigated by the circumstance that some observations on ferrite were made with high-temperature microscopy.

Brown *et al.*[14] found that proeutectoid ferrite precipitating on austenite twin boundaries in Fe-C-Mo is "clean," *i.e.*, it is relatively dislocation-free. On the other hand, ferrite formed on austenite grain boundaries is wrinkled on one side and clean on the other side. (The term "wrinkled ferrite" was introduced by Hultgren[15] on the basis of optical metallographic studies of ferrite and bainite formation in ternary and higher order Fe-C-X alloys.) Ferrite formed on austenite grain boundaries is wrinkled on one side (growth into the non-Kurdjumov-Sachs [K-S][16] oriented austenite grain) and clean on the other side (presumably, growth into the K-S oriented austenite grain). Wrinkled ferrite growth interfaces tend to be ragged, and, at later stages of ferrite growth, sympathetic nucleation appears to dominate. These important observations likely derive from the compensation of misfit across a K-S interface by a combination of single sets of misfit dislocations and of structural disconnections.[17,18,19] (The term "disconnection" is currently applied to ledges also having a dislocation character.[20,21]) At non-K-S interfaces, the misfit compensation geometry is evidently more complicated. Furuhara and Maki[22] made a TEM study of bcc grain boundary ferrite allotriomorphs formed in the fcc matrix of a Ni-45 wt. pct Cr alloy. As in the prior study of allotriomorph interfaces in a Ti-7 pct Cr alloy,[23] both nearly or exactly rationally and irrationally oriented interfaces were partly coherent. In one of their micrographs (Figure 2(a)), in which appreciable amounts of fcc matrix were shown on both sides of grain boundary allotriomorphs, somewhat more dislocations appeared to be associated with the non-Nishiyama[24]-Wassermann[25] interface than with the (exactly) N-W interface. This suggests the possibility that the additional dislocations played a role in the compensation of misfit across the non-Nishiyama-Wassermann interfaces. When these interfaces are overrun by growth ledges, such "excess" dislocations may be rejected into the ferrite phase.

Yanar *et al.*[26] observed microtwins, twins and split dislocations in the τ_m phase of MnAl(C) alloys. They also showed that a sharply curved edge of an ε:τ_m interface is resolved under high-resolution TEM into a succession of closely spaced ledges three atomic layers high.[27] The intersections of the terraces and risers of these ledges may have served as lines of concentrated stress in the presence of volume and shape changes and hence as possible sites for dislocation nucleation. The possibility of their crystallographic participation in twin formation will be considered later.

The salient features of the preceding observations are summarized in Table I, and are schematically illustrated in Figure 1.

III. EXPLANATIONS FOR ORIGINS OF INTERNAL STRUCTURE

The accommodation of changes in volume per atom during the MT is the traditional explanation for the evolution of internal structure.[1] If we liken an MT to the formation of a precipitate in a solid, then we must add to this effect those associated with the shape change involving a change in stacking sequence, even in diffusional phase transformations.[30] Additionally, both elastic and plastic incompatibilities at the parent:MT interfaces may have to be accommodated with dislocations and/or twins in one or both of the participating phases.

We envisage that slip dislocations observed in the matrix ahead of the parent:MT interfaces in Cu-Ga[2] and Ag-Al alloys[3,4] may arise at least in part due to one or more of the foregoing sources. For the sake of discussion, let us consider a case where the average volume per atom of the massive phase is larger than that of the parent phase. This situation would impose compressive stresses on the adjoining matrix. These stresses would be relieved if vacancy-type prismatic loops were injected into the matrix when it is softer than the massive phase. Dislocations associated with the massive:matrix boundary may be involved in the formation of these loops. For example, an interfacial dislocation with Burgers vector $\mathbf{b}_i$ can dissociate into a matrix dislocation with Burgers vector $\mathbf{b}_m$ according to the following reaction:

$$\mathbf{b}_i \rightarrow \mathbf{b}_m + \mathbf{b}_r$$

where $\mathbf{b}_r$ is the Burgers vector of the residual dislocation left at the interface. The matrix dislocations thus formed can then multiply as envisaged by Ashby and Johnson,[31] resulting in development of prismatic loops. Furthermore, the interfacial dislocation reforms after the generation of each loop.

Alternately, the matrix dislocations in this reaction could behave as a Frank-Read source since their ends are pinned at the massive:matrix boundary. However, to accommodate fully the volume change, it is necessary to create many Frank-Read sources along the interface. Although incoherent boundaries should provide a higher number density of these sources, partly coherent boundaries may be able to furnish a reasonable number of them at the risers of growth and other disconnections. At these sites, dislocations on the risers may be pinned where they meet the bounding terraces and can expand under the impetus of volume and shape change stresses into either the matrix or the product phase. Boundaries where edge-to-edge partial coherency exists instead of plane-to-plane coherency may also be fruitful in this regard because they often appear to lie in either shallower energy

Table I. Experimental Observations on Internal Structure of Massive Transformation Products

Alloy	Massive Transformation	Crystal Structure Change	Observed Internal Structures
Cu-(19.5 to 20.5) pct Ga[6,28,29]	$\beta \rightarrow \alpha_m$	bcc $\rightarrow$ fcc	dislocations slightly split*
Cu-(20.5 to 25.5) pct Ga[6,28,29]	$\beta \rightarrow \zeta_m$	bcc $\rightarrow$ hcp	profuse twinning, widely split dislocations
Cu-37.6 Pct Zn[7]	$\beta \rightarrow \alpha_m$	bcc $\rightarrow$ fcc	large twins and microtwins, high densities of aligned dislocations
Ag-24.5 Pct Al[3,4]	$\beta \rightarrow \zeta_m$	bcc $\rightarrow$ hcp	twins and well split dislocations
Fe[8–11]	$\gamma \rightarrow \alpha_m$	fcc $\rightarrow$ bcc	dislocations only
MnAl(C)[26]	$\varepsilon \rightarrow \tau_m$	hcp $\rightarrow$ $L1_o$	twins, microtwins, split dislocations

*The α_m/ζ_m duplex structure is not considered here.

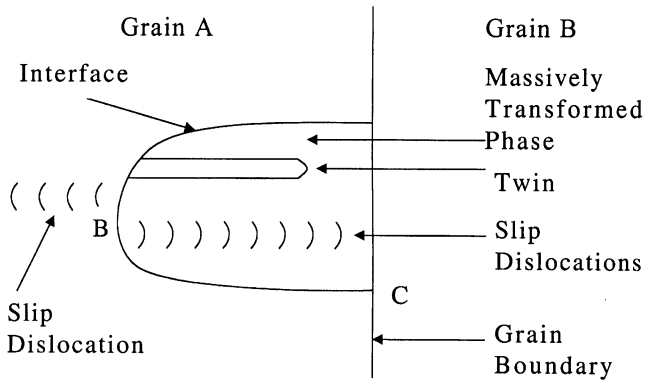

Fig. 1—Schematic of microstructural features in and around a phase produced by a massive transformation.

cusps or outside such boundary orientations.[32,33] If massive:matrix interfaces are incoherent,[1,34,35] coherency stresses are absent. On the view that these interfaces are partly coherent,[33,36] these stresses could play a role in initiating plastic deformation at massive:matrix interfaces, as implied by Wilson and Chong.[37] However, these stresses would be important only when interfaces are fully coherent, a situation obtained only during the very earliest (and so far unobservable) stages of growth fcc $\leftrightarrow$ bcc and bcc $\leftrightarrow$ hcp transformations in bulk alloys.[38] The relief of these stresses is then quickly accomplished by the acquisition of suitable linear misfit compensating defects. Full coherency appears to exist for perceptible intervals of reaction time only in fcc $\leftrightarrow$ hcp transformations, and then only in the presence of their standard low energy orientation relationship and conjugate habit planes, and also when misfit is minimal across these planes.[39]

In the context of edge-to-edge matching, originally proposed by Zhang and Kelly,[40,41] Nie and Muddle[32] and Howe *et al.*[42] have presented experimental evidence supporting the presence of such interfaces during the $\alpha \rightarrow \gamma_m$ transformation in a near-TiAl alloy. Unlike plane-to-plane matching, pairs of planes matching at their edges can climb in quasi-independent fashion.[33] The suggestion is now offered that this climbing process could lead directly to point defect emission and to dislocation formation. Further, these defects will always appear in the product phase.

Gleiter,[43] Gleiter *et al.*,[44] and Mahajan *et al.*[45] have suggested that migrating grain boundaries could be the source of dislocations and twins in materials due to "growth accidents." These boundaries sometimes have low energy facets. When the boundaries move during grain growth, mistakes in stacking sequence can occur during the migration of these facets. The higher the velocity of the boundaries, the higher the probability of such mistakes. This linkage is experimentally supported by observations on the increasing density of dislocations and twins produced at the migrating boundary in an InP bicrystal with increasing migration rate.[44]

In the following, we apply the concept of "growth accidents" to explain the formation of internal substructure observed in MT phases. We liken a massive:matrix interface to a grain boundary consisting of low energy facets and ledges. The observations summarized in Table I show that both dislocations, usually more or less well split or dissociated, and twins are present when the product phase is fcc, hcp or $L1_o$ (fcc-related tetragonal). Stacking fault energies in the fcc and hcp phases tend to be low. Since the high-purity solvent metals, Cu and Ag, have a moderate to low stacking fault energy,[46] faults in their terminal solid solution alloys doubtless have still lower energies. Dissociation of dislocations in τ_m MnAl(C) indicates that the stacking fault energy in this material must also be low. Among the alloys just noted, twins appear to be absent only in α_m Cu-Ga. This absence may be only apparent since the composition range of this MT is heavily overlapped by that of ζ_m formation, resulting in domination of the microstructure by a "feathery structure" developed by the simultaneous formation of α_m and ζ_m as alternating lamellae.[28]

Now consider a situation where a {111} facet is present on the massive:matrix interface ABC shown in Figure 1, and the material is undergoing a bcc $\rightarrow$ fcc$_m$ transformation. If "growth mistakes" occur on the {111} plane during this transformation, then we can rationalize the origins of stacking faults that are bounded by Shockley partials and twins in terms of the arguments of Gleiter,[43] Gleiter *et al.*,[44] and Mahajan *et al.*[45] Their argument for "growth accident" development will first be summarized for grain boundary migration. Layers of growth accident are taken to have formed by two-dimensional nucleation (for which the driving force available may have been inadequate for detectable kinetics) or by the winding up of a screw dislocation. The "accident" itself results when an atom plane is added containing "a row of atoms displaced from the sites they would occupy in the perfect lattice." This results in empty channels too narrow to accommodate another row of atoms. If the next atom plane resumes the original stacking sequence, a pair of very closely spaced Shockley partials results.

Since the product phase in the MTs described in Table I is fcc, near-fcc or hcp in five of the six transformations considered, this mechanism translates directly into the present problem, where the development of a dislocation which will move away from the interphase boundary (instead of a grain boundary) must be considered. When the massive:matrix boundary advances more rapidly than the "growth accident" can climb or glide, the "accident" will be trapped in this phase. However, in the reverse situation, the "accident" will move into one of the phases forming the boundary provided that it can do so by glide. Since the growth kinetics of massive transformations are generally accepted as controlled by trans-interphase boundary diffusion, dislocation climb by volume diffusion would be too slow to permit outrunning the massive:matrix interface—unless the number density of growth ledges in the interface is small. The end points of the trapped dislocation are anchored at the interphase boundary. Hence loops of varying sizes appear, depending upon when an individual massive crystal was nucleated as well as the structural details of its growth kinetics. These loops can interact and thereby become no longer distinguishable.

Mahajan *et al.*[45] have proposed two somewhat similar mechanisms through which annealing twins can also form from growth accidents in fcc metals. If a curved grain boundary consists of a sequence of monatomic ledges with {111} facets at the atomic scale, growth accidents on these facets will result in pairs of Shockley partials on the terraces of these ledges. Through repulsion between these partials, microtwins will propagate into the grain containing the ledges. These will consolidate into thicker or macrotwins only when a long and unbroken sequence of monatomic ledges is present. Their second mechanism, following that of Gleiter,[43] is based upon areas of a grain boundary lying in a {111} plane. As the grain boundary moves normal to this plane (by a mechanism not specified), the growth accident on the {111} plane will develop into an annealing twin by the glide of Shockley partials as envisaged by Mahajan *et al.*[45]

Applying these considerations to the massive transformation, on the view that massive:matrix interfaces are partly or fully coherent,[33,36] when the terrace plane of structural[17,18,19] and/or of misfit-compensating ledges[5,47,48] or disconnections[20,21] is parallel to the habit plane of a twin these planes should also provide sites for "growth accidents." These accidents may occur during the passage of successive growth ledges (probably growth disconnections in most cases)—whose terrace plane may itself contain disconnections—over the static disconnections. Particularly in the case of structural disconnections, whose height is usually only a few interatomic spacings, growth accidents on a succession of their terraces would lead to twinning by the first mechanism of Mahajan *et al.*[45] As in the case of annealing twin formation during grain growth, the previously noted observation by Yanar *et al.*[27] of closely spaced ledges three atomic layers high at a sharply curved edge of an ε:τ_m boundary in MnAl(C) would provide a suitable "template" for the formation of a succession of microtwins, provided that the terrace planes are suitable sites for "growth accidents."

The previously noted observations of Malcolm and Purdy[7] that β:α_m interfaces formed during quenching a Cu-Zn alloy could generate dislocations within α_m but that α of more nearly equilibrium composition subsequently formed result in a nearly dislocation-free product may be explained as follows. Dislocations probably are generated by the migrating massive:matrix interfaces, but they subsequently undergo self-glide and climb during isothermal growth of near-equilibrium α, resulting in annihilation and rearrangement of these dislocations.

The $\gamma \rightarrow \alpha_m$ transformation in iron occurs at fairly high temperatures. Since annealing twins are not observed in bcc iron, implying a high stacking fault energy, and since deformation twinning occurs in this metal only at low temperatures, it is not surprising that only dislocations are seen in α_m. These dislocations could form due to "growth accidents" as well as plastic deformation induced by volume and shape changes.

The various mechanisms considered for both dislocation and twin formation should all tend to operate more rapidly the faster the migration of interphase boundaries. This is consistent with formation of these defects with greater frequency the larger the driving force for the massive transformation and the proeutectoid ferrite reaction in an Fe-C-Mo alloy.[2,3,4,7,49]

A reviewer has brought to our attention papers by Yanar *et al.*[51,52] in which the mechanism of Mahajan *et al.*[45] has been applied to twin formation during the $\varepsilon \rightarrow \tau_m$ transformation in MnAl(C). However, the origins of slip in the product phase, for which this mechanism can also account, were not considered and the possibilities of applying this mechanism to transformations in diverse other alloy systems and to precipitation from solid solution were not discussed.

IV. CONCLUSIONS

Observations made on the internal structure developed during six massive transformations are summarized from the literature. These transformations include $\beta \rightarrow \alpha_m$ Cu-Ga, $\beta \rightarrow \zeta_m$ Cu-Ga, $\beta \rightarrow \alpha_m$ Cu-Zn, $\beta \rightarrow \zeta_m$ Ag-Al, $\varepsilon \rightarrow \tau_m$ MnAl(C) and $\gamma \rightarrow \alpha$ Fe. The following conclusions were drawn from these observations.

1. Except in the case of $\gamma \rightarrow \alpha_m$ Fe and with the possible exception of $\beta \rightarrow \alpha_m$ Cu-Ga, the internal structure of the massive product includes both twins and slip. The apparently low stacking fault energy of the product phases and the doubtless high stacking fault energy in α Fe appear to be responsible for these differences.
2. Although plastic relaxation of shape changes as well as volume changes presumably contributes to the observed internal structures, the circumstance that the product phase is harder than the matrix phase in at least four of the six transformations considered indicates that this cannot be the sole factor responsible for such structures. Further evidence that that these are not the only factors responsible for dislocation generation during the MT is provided by the appearance of such dislocations during hot-stage TEM studies, where the constraints causing plastic deformation are reduced in the thin foils used.[50]
3. Coherency stresses are unlikely to contribute significantly to the internal structures unless misfit compensating defects are slow to form at massive:matrix boundaries. These stresses are relieved, usually quite rapidly, by formation of linear misfit compensating defects at their interphase boundaries.
4. Hot-stage TEM observations of dislocation emission from interphase boundaries during the $\beta \rightarrow \zeta_m$ transformation in Ag-24.5 pct Al[3,4] have been considered on

the basis of studies on dislocation and twin formation at moving grain boundaries.[44,45] Dislocation emission in ζ_m may occur by means of "growth accidents." These are defined[44,45] as Shockley partial pairs formed by stacking errors. When β:ζ_m boundaries move faster than the "growth accidents" can climb, the "accidents" are trapped, and may move into ζ_m as dislocation loops. Twin formation in ζ_m can occur at growth ledges on β:ζ_m boundaries.

5. Misfit dislocations at massive:matrix boundaries as well as those on the risers of disconnections connecting their terraces may serve as Frank-Reed sources for loops extending into either phase.
6. Massive:matrix boundaries formed by edge-to-edge[40,41] matching, as in the $\alpha \rightarrow \gamma_m$ transformation in a near-TiAl alloy,[32] can migrate by climb and thereby introduce dislocations and loops thereof in the massive phase.
7. "Wrinkled ferrite," given this appearance by the presence of appreciable densities of dislocations produced during proeutectoid ferrite formation in Fe-C-Mo and other steels,[15] is associated with interfaces formed by a non-K-S[16] orientation relationship.[14] Ferrite bearing a more or less exactly K-S or other rational orientation relationship has a much lower dislocation density.[14] The emission of dislocations from the former interfaces appears to be associated with a particularly complex interfacial structure[22] whose overrunning by growth ledges may release "excess" dislocations into the product phase.

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