The Nature and Role of Incoherent Interphase Interfaces in Diffusional Solid-Solid Phase Transformations

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In this article, some views on the nature of incoherent interphase interfaces, and their role in the nucleation and growth processes governing the evolution of microstructure in solid-state diffusional transformations (reconstructive transformations), are explored. It is argued that essentially incoherent interfaces can be involved in the initiation and propagation of polymorphic transformations and massive transformations as well as in various precipitation phenomena in metallic and ceramic systems. Similar views have already been advanced earlier in connection with studies of massive transformations. Faceting along the interphase interface during nucleation and growth can derive from thermodynamic, kinetic, and crystallographic factors independent of the bicrystallography of the conjugate phases. This idiomorphic behavior can be relevant to both intergranular and intragranular phase formation. The concept of one-dimensional (1-D) commensuration of phases through plane edge-to-edge/row matching is an interesting extension of the classic ideas of coherency and bicrystallography and potentially important in characterizing the behavior of certain types of boundaries. However, the general importance of these geometrical relations in real and reciprocal space will depend on the depth of the energy wells in orientation space associated with these special boundaries.

PREAMBLE

This Symposium has been organized within the framework of a series of symposia honoring the winners of the annual TMS Hume-Rothery award, and in commemoration of the many seminal contributions to the understanding of the stability of metals and alloys made by the late Professor Hume-Rothery. On the occasion of this symposium, we wish to congratulate Professor Hub Aaronson, the current Hume-Rothery Award winner, for his many contributions to the field of phase transformations.

One of the authors of this paper (TBM) had known Hume-Rothery over many years. Perhaps a few brief remarks about Professor Hume-Rothery's life and achievements are in order. William Hume-Rothery was born in 1899 and spent almost all of his scientific career as a Royal Society Fellow, and later as Professor and Head of Department in the discipline of Metallurgy/Materials in Oxford. During his early boyhood, he was originally destined to follow a military career, but at age 18, he contracted cerebral spinal meningitis that left him totally deaf. This event turned Hume-Rothery toward a scientific carrier of a life-long research into the properties of metals and alloys. He managed to contribute many outstanding ideas that have made a permanent impact on the field of materials science.[1,2]

Despite his tremendous hearing handicap, Hume-Rothery lectured well without being able to hear a single spoken word. To accomplish this, he would invite an associate to sit in the front row and give him hand signals indicating the level and pitch of his voice. His many statements and approaches to controversial issues, or disagreements, are well known to his colleagues. His attitude may well be paraphrased as follows: ". . . if there are a number of experimental facts, and if even one of them does not fit into the existing theory, then reject the theory, or modify it to fit all the facts." This motto seems particularly fitting to the topic and the contents to this symposium.

I. INTRODUCTION

THE present contribution has originated from an initial invitation by Professor Aaronson to have a paper in the symposium that would be presented ". . . as the last talk in which . . . the incoherency view on the structure of irrational interface boundaries would be represented . . .".

It is very satisfying to us that the "incoherency view" has been supported and embraced by numerous contributions to this symposium from differing points of view. Importantly, this forum has revealed a rich diversity in the nature and behavior of interphase interfaces involved in solid-state transformations. This focus has yielded a number of new concepts and ideas that should broaden our perspectives on the role of incoherent boundaries.

The nature and behavior of interphase interfaces in materials represents a critical area in the development of a basic understanding of the evolution of microstructures during processing and heat treatment. The structure and properties of the interphase interfaces are fundamental to the basic processes of nucleation and growth involved in phase transformations. Generally, the behavior of the transformation fronts ultimately governs the resultant morphology of the transformation products and therefore the resultant physical and mechanical properties of an engineered material. Howe's book on Interfaces in Materials[3] dealing with the atomic

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This article is based on a presentation made in the "Hume-Rothery Symposium on Structure and Diffusional Growth Mechanisms of Irrational Interphase Boundaries," which occurred during the TMS Winter meeting, March 15–17, 2004, in Charlotte, NC, under the auspices of the TMS Alloy Phases Committee and the co-sponsorship of the TMS-ASM Phase Transformations Committee.

structure and the thermodynamics and kinetics of various interfacial phenomena in materials provides a modern perspective of this subject. The subject of interphase interfaces and their role in solid-solid transformations was hotly debated during a recent international symposium held in St. Louis in fall 2000, "The Mechanisms of the Massive Transformation." The proceedings of this conference were published in *Metallurgical and Materials Transactions* in August 2002. Many of the issues addressed in this Hume-Rothery Award Symposium honoring Professor H.I. Aaronson derive directly or indirectly from the presentations and discussions at the St. Louis meeting. A particularly controversial subject has arisen regarding the role of coherency and bicrystallographic matching between parent and product phases and the frequent occurrence of facets on the interface propagating the massive product. The discussion brought into question the very definition of a "massive transformation" as a distinct mode of solid-state phase transformation occurring in metallic and ceramic systems. Of course, Professor Aaronson's interests and contributions in the area of interphase interfaces extend far beyond the massive transformation as this Hume-Rothery Symposium attests.

Over the past several years, an important concept has arisen regarding the extension of the concept of coherency with emphasis on plane edge-to-edge/atom row matching across interfaces, as discussed by a number of investigators.[4–9] Kelly and Zhang[7] have emphasized the importance of correspondence of relatively close-packed rows of atoms at the edges of the planes. In this contribution, the focus will be on the role of incoherent interphase interfaces in the initiation and propagation of polymorphic and massive transformations occurring in the solid state, that is, thermodynamically first-order phase transformations involving a change in the crystal structure without attendant change in composition. An important thesis in this article is that relatively low energy incoherent interfaces can arise during nucleation and growth as a result of preferential formation of low energy interfaces specific to the emerging phase (energetically and kinetically). In this context, an incoherent interphase boundary is taken to be one where atomic matching at the interface is minimal and across which generally no essential, rational crystallographic relationships exist. The occurrence of one-dimensional commensuration between the conjugate phases producing plane edge-to-edge or atom row matching is regarded as a semicoherent boundary or a basically incoherent boundary punctuated by a periodic matching of specific planes or atomic rows. Howe *et al.*[10] have attempted a systematic classification of interphase boundary structures in crystalline solids including a discussion of types of incoherent interfaces. They call attention to the case where the orientation relationship (OR) between the conjugate phases is irrational and the apparent habit plane is low index in only one phase (type-3 incoherent). They suggest that this type of boundary will exhibit a reduced energy compared to a boundary with an irrational OR and no rational planes in the pairs comprising the plane of contact/conjugate habit planes. This is identical to the special type of incoherent boundary mentioned previously and of special interest in this discourse.

In recent discussions of the role of the interphase interfaces in solid-state transformations, the terms *bicrystallography* and *monocrystallography* have been used to describe the relationship between various aspects of coexisting phases. Bicrystallography refers to the conditions at an interphase interface resulting from characteristic atomic matching and lattice correspondences across the contact region, and this may involve correspondence of unit cells, planes, or directions, including the existence of invariant lines or edge-to-edge matching of planes. The term monocrystallography has been applied to conditions at the contact region between phases that are largely determined by the growth kinetics and natural habit of planes associated with the emerging phase and that are not substantially perturbed by the crystallography of the surrounding matrix phase. In this situation, the matrix is primarily a chemical potential reservoir that provides the "driving force" for the phase transition. In the crystal growth literature, the emergence of crystalline phases, which display characteristic faces and morphologies indicative of their underlying symmetry, is a ubiquitous phenomenon, and these growth topographies are called *euhedral* or *idiomorphic*. Minerologists refer to microconstituents of a rock as idiomorphic when these microconstituents exhibit their own characteristic morphology or form and not one forced upon them by the other constituents—that is, they tend to display their own characteristic form or habit. Hillert,[11] in discussions of bicrystallographic and monocrystallographic interfacial relations, has suggested the terms *diamorphic* and *idiomorphic*. The *dia-* and *idio-* prefixes refer to a communication across the contact region between phases and the lack of essential dialogue or interactions, respectively. In solid-solid transformations, idiomorphic behavior might be expected where chemical bonding and strong anisotropy in the surface energy dominate bicrystallographic/diamorphic factors at the plane of contact. The morphology of the emerging phase is thus primarily determined by the specificities of atomic attachment to the growing crystal and its intrinsic surface energetics. The crystalline forms that develop during growth generally are kinetically determined rather than equilibrium shapes, as imposed by the Wulff construction.

In this article, the authors discuss the role of interphase boundary structure and energetics in the evolution of morphology in diffusional phase transformations during nucleation and growth with particular attention focused on the idiomorphic behavior of the growing phase. The term diffusional here refers to a *reconstructive transformation*, according to Buerger,[12] whereby a new atomic or molecular configuration is assembled by essentially uncoordinated, thermally activated atomic migration. Such a category of transformation has been called "civilian" by Christian[13] as opposed to the "military" transformations specified by Frank,[14] which are effected by coordinated or synchronous motions of groups of atoms such as in the case of martensitic transformations. The diffusional or reconstructive category is taken to include trans-boundary atomic migration generally associated with the massive transformation and certain polymorphic phase changes.[15]

II. NUCLEATION

The discussion of nucleation and growth processes with emphasis on the nature of the interphase interface that emerges between the parent and product phases calls attention to a quote from J.W. Christian's treatise, "The Theory of Transformations in Metals and Alloys: Parts I and II":

> ". . . , the Wulff thermodynamic criterion is applicable only to nuclei or tiny crystallites which have surface energies comparable to their bulk free energies. The shape of a macroscopic crystal is governed in practice by the growth rates of the various possible faces[16]".

The quote from Christian's classic work was taken from the context of a discussion of crystal growth from the vapor phase and the applicability of the Volmer–Weber–Becker–Doering classical nucleation formalism. However, the same point is emphasized here, that is, the energetics of the interphase interface and the anisotropy of the interfacial free energy will be paramount during the nucleation stages in solid-solid transformations, while the subsequent behavior during growth of the transformation front will be dictated primarily by kinetic factors. Of course, the interface structure enveloping the initial nucleus must be inherited to some extent during subsequent growth and can play a central role in the kinetic behavior and evolution of the interface topography. For example, the structural constraints of coherency or semicoherency (including one-dimensional (1-D) commensuration) can profoundly influence the possible atomistic mechanisms that effect the migration of the interphase boundary normal to itself.[3,10]

There has persisted over many years a school of thought that essentially suggests that incoherent interphase interfaces cannot play a role in the nucleation (homogeneous or heterogeneous) of a new phase in solid-solid transformations because of the need to minimize the surface term in the energetics of nucleation. Indeed, in the Pittsburgh symposium in 1980, Plichta *et al.*[17] strongly supported the view expressed earlier[18] that ". . . special crystallographic orientation relationships do indeed obtain in massive transformations." It was proposed that such ". . . special crystallography develops during nucleation and is an inherent characteristic of massive transformations in general. Once established during nucleation, . . . low energy boundaries initially fully coherent can be expected to become partially coherent during growth". Our discussion is aimed at questioning this categorical exclusion of any role of ostensibly incoherent interphase boundaries in the nucleation (and growth) processes, which are ubiquitous in solid-solid transformations. We begin by considering the heterogeneous nucleation of a new phase on a grain boundary face according to a classic model[19] depicted in Figure 1(a). It is assumed that the nucleus forms a low energy coherent or semicoherent interface with one grain and an incoherent interface with the other. The probability of the nucleus establishing coherency with respect to both grains in a randomly oriented polycrystalline aggregate is expected to be low. A variant of this simple grain boundary nucleus model, which can effect an important reduction in the total interfacial free energy of the nucleus, is for the incoherent α-β boundary section to facet, as depicted in Figure 1(b). Importantly, the facets can exhibit low index habit planes characteristic of the nucleus, that is, exhibiting intrinsic symmetries of the new phase independent of essential crystallographic matching with the matrix across the interphase interface, as mentioned in the Introduction. This idiomorphic behavior can be expected to lower the effective interfacial free energy and thus decrease the nucleation barrier, ΔG^*. The early stages of nucleation and

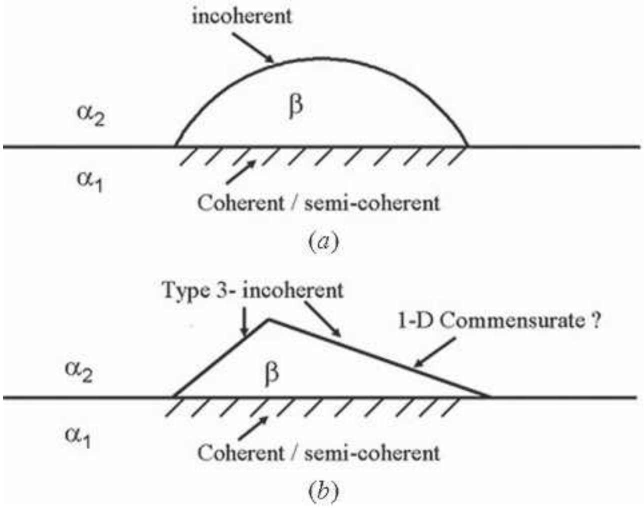

Fig. 1—(*a*) Classical critical nucleus model for singly faceted grain boundary nucleus, which is incoherent with respect to grain α_2 but coherent or semicoherent with respect to α_1. (*b*) Grain boundary nucleus involving faceting of the incoherent boundary segment.

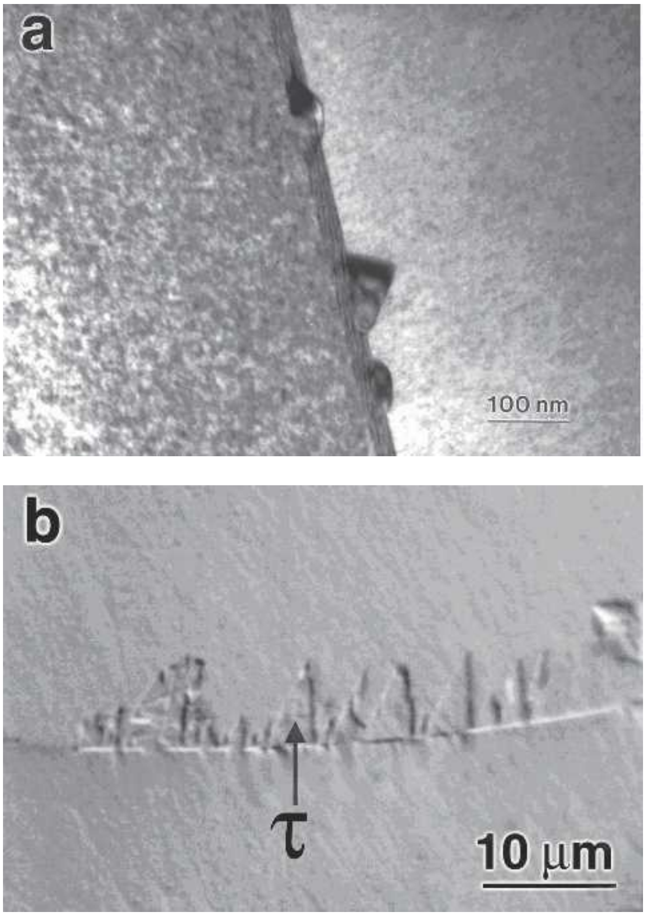

Fig. 2—(*a*) TEM showing grain boundary nucleation and growth of L1o τ phase in Mn-Al-(C) alloy during massive transformation $\varepsilon\,(\mathrm{hcp}) \rightarrow \mathrm{L1o}$. (*b*) Optical micrograph of faceted grain boundary idiomorphs with Burgers OR with respect to one grain and apparent incoherent boundary with respect to the other. Growth is restricted along the semicoherent boundary segment with the Burgers OR but occurs readily into adjacent grain (20).

growth captured in the micrographs of Figures 2(a) and (b), revealing faceted morphologies characteristic of the massive transformation giving rise to the τ phase ($L1_o$) in Mn-Al-(C) permanent magnet alloys, are indicative of this phenomenon. The grain boundary τ-phase particles appear to have no rational orientation relationship with respect to the grain into which they are growing preferentially and typically exhibit faceted or serrated boundary topographies. The relatively immobile interface at the grain boundary has been shown to be semicoherent fitting the classic picture rather well.[20] A variation of this process could involve bicrystallography in that a low energy planar boundary segment could occur through 1-D commensuration or plane edge-to-edge matching, as shown in Figure 1(b). Thus, a so-called Δg interface, or Moire′ plane,[8,9] is formed along a segment of the α_2-β interphase interface, which also can reduce the overall interfacial free energy expenditure in the nucleation process. Even relatively small reductions in the interfacial free energy can be significant because of the strong functional dependence of ΔG^* on the interfacial free energy according to the classical nucleation theory. It must be emphasized, however, that the classic variant depicted in Figure 1(a) is a viable nucleus ($\Delta G^* < 60\ k_B T$) depending on the magnitude of the various interfacial free energies involved and the driving force in any specific system.[16,21]

It is important to point out that the preceding analysis is relevant to grain edge and grain corner nucleation.[22] Also, it is possible that intragranular idiomorphs may arise, and indeed, there is experimental evidence for such behavior.[23] Clearly, both monocrystallographic/idiomorphic and bicrystallographic behavior are expected to occur in solid-solid transformations.

III. GROWTH

The structure of the interphase interface exerts a major influence on the mechanisms involved in the propagation of a transformation front during growth. As mentioned in Section II, coherency (1-D or two-dimensional (2-D)) generally requires a ledge mechanism to effect the growth of the new phase into the parent phase or matrix without extraordinary atomic rearrangements within the interfacial region.[3,10] It has been established experimentally that incoherent interfaces can be smoothly curved or faceted on the mesoscale and that apparently smoothly curved interface regions might be faceted on smaller length scales down to the nano- and atomic scales,[20] *e.g.*, Figures 3 and 4. These migrating disordered, incoherent boundaries have been observed to readily cross many matrix grain boundaries in some massive transformations and in some cases produce single crystals from polycrystalline aggregates.[15]

In the analysis of the mechanisms of growth controlled by thermally activated atomic jumping across the interphase interfaces (trans-boundary diffusion), the question often arises whether the atomic detachment and attachment processes are effectively a quasi-continuous, nearly random jumping of individual atoms from the parent to the product phase, or basically a ledge growth mechanism controlled by successive nucleation and lateral movement of ledges. The continuous growth mechanism of random attachment is essentially associated with a relatively "rough" interface, whereas the ledge mechanism involves "smooth" terraces punctuated by ledges and kinks on length scales dictated by the characteristic ledge height, *h*, and ledge spacing, λ. It has been suggested that effective control of interface migration by a ledge mechanism is physically ambiguous when λ/*h* is less than an order of magnitude.[24] Recent kinetic studies of massive transformations in Ti-Al[25] and Mn-Al-C[20] alloys indicate that the growth rates are controlled essentially by a quasi-continuous process, as deduced from analysis based on a modified Burke–Turnbull equation. This is consistent with HREM (High Resolution Electron Microscopy) observations that apparently flat or smooth interphase interfaces in the Mn-Al-C system are often faceted on the atomic scale, effectively producing rough interfaces (Figure 3).

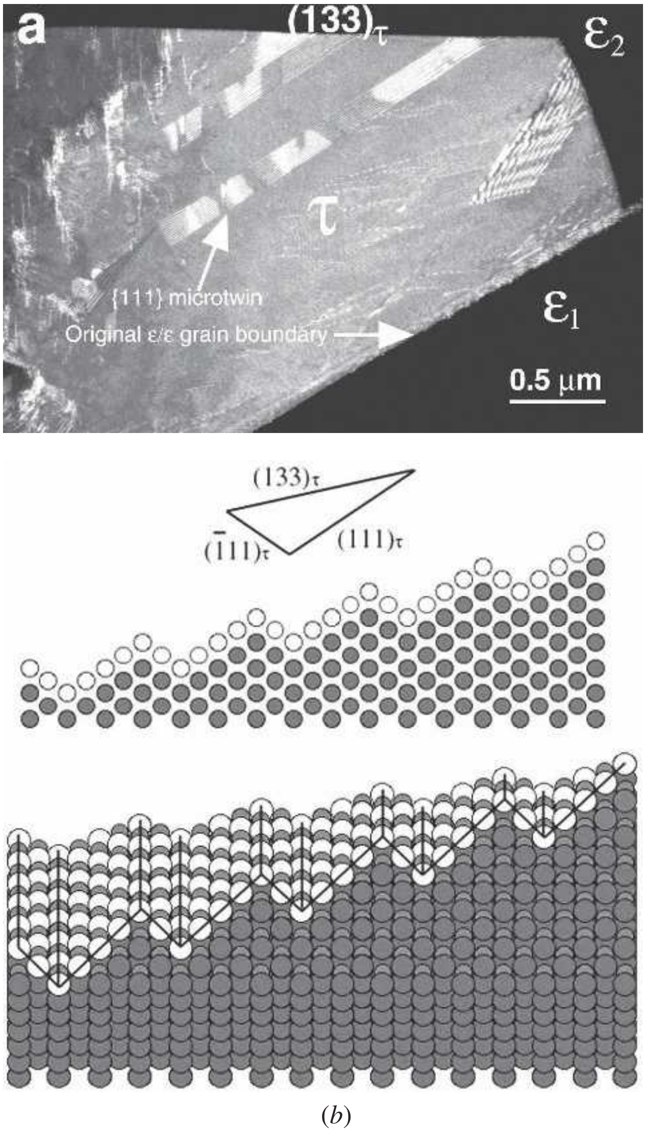

Fig. 3—(*a*) Prominent $(133)_\tau$ planar facet at the ε-τ interface in Mn-Al-(C) alloy during massive growth into grain ε_2. (*b*) Schematic of a {111} nanofaceted interface producing an apparent $(133)_\tau$ high-index plane (20).

Important questions in the discussion of the growth processes and faceting are as follows.

1. Is faceting on any length scale necessarily indicative of some bicrystallographic matching/coherency (1-D or 2-D)?
2. Can an incoherent interphase interface exhibiting no systematic OR with respect to the matrix phase into which it is growing and apparently no characteristic atomic

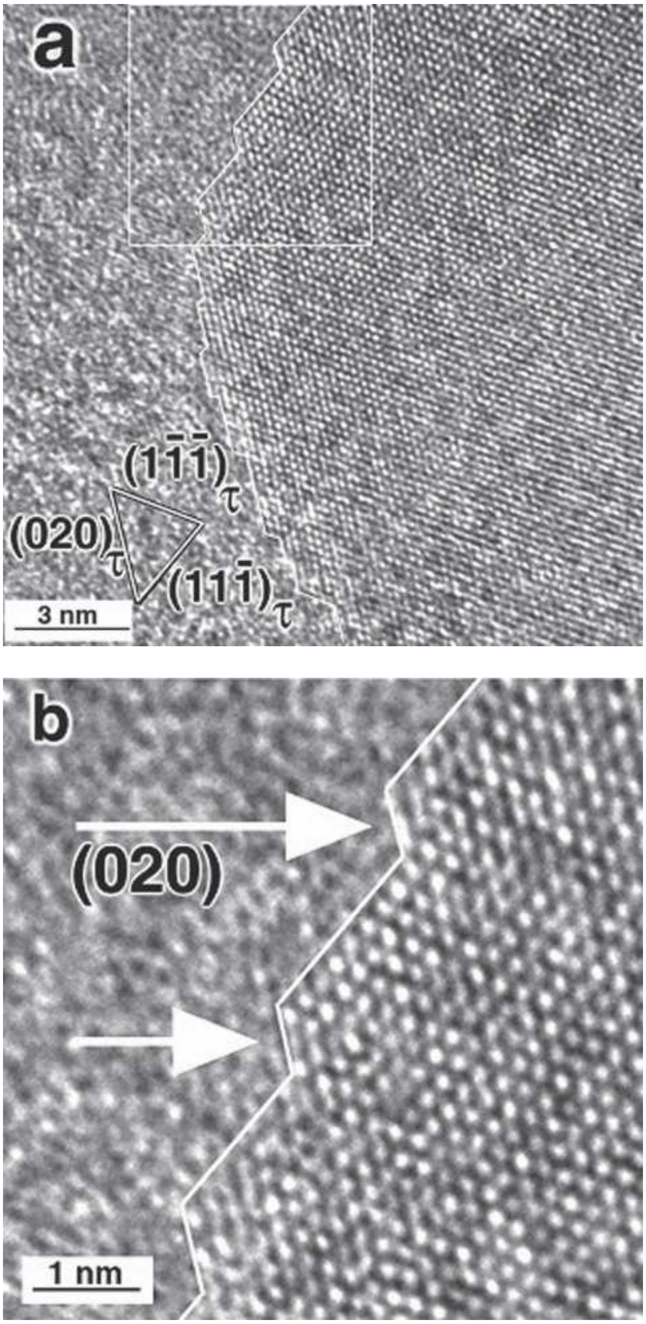

Fig. 4—(*a*) HREM image of a curved section of an ε-τ interface and (*b*) higher magnification of selected area showing composite (020) and (111) nanofacets/terraces/ledges (20).

matching between the product and parent phase exhibit prominent and persistent planar sections along the migrating boundary?

(3) Do both possibilities occur in the evolution of the interphase boundary topography depending on the structural and energetic specificities of the conjugate phases?

It is argued here that the behavior associated with question (2) derives directly from what has been called idiomorphic growth in preceding sections I, II, and III, that is, a highly faceted interphase boundary can arise during growth stemming primarily from the nature of the emerging phase. Idiomorphic growth occurs when the interfacial energetics and atomic attachment processes are dominated by the structure and symmetry of the growing phase allowing the phase to display to varying degrees its "natural" habit within the surrounding matrix. During transformation, this growth morphology is likely to be a kinetically determined shape rather than an equilibrium shape dictated by the Wulff construction. This idiomorphic growth might be expected to prevail in cases where the interfacial energy and structure show strong anisotropies associated with preferred bonding coordinations within specific crystallographic planes, *e.g.*, in Zr-N.[26] Yanar *et al*.[20] have suggested that in the growth of the intermetallic MnAl($L1_o$) phase during massive transformation in the Mn-Al-(C) system, the migration of the interfaces of mixed composition is different than those comprised of atomically unmixed layers. It is asserted in the present discussion that faceting along an interphase boundary during growth can occur on various length scales (meso → nano → atomic) with characteristic crystallographic features independent of bicrystallographic or atomic matching between the parent and product phases and devoid of any characteristic linear-misfit defect structure; that is, in some instances, electronic and bonding considerations may dominate crystal geometry. However, faceting of apparent incoherent interphase boundaries during transformation can occur if plane edge-to-edge/row matching is established, producing energy wells in orientation space and interfacial structures, which inhibit migration. The so-called Moiré or $\Delta\mathbf{g}$ planes defining the crystallographic orientation of the boundary and defect structure are expected to require ledgewise growth (Moiré ledges) within the boundary plane constraining boundary mobility.[9]

It has been mentioned several times that faceting on incoherent interphase boundaries has been observed to occur on various length scales. The HREM studies have indicated that broad planar sections on the mesoscale showing an apparent high index habit plane can be nanofaceted along characteristic low index planes. Also, planar sections on various length scales can lead to twin formation *via* 2-D nucleation on the migrating interface.[20]

IV. DISCUSSION

This 2004 Hume-Rothery Award Symposium focusing on the "Structure and Diffusional Growth Mechanisms of Irrational Interphase Boundaries" apparently has embraced the idea of so-called type-3 incoherent or irrational interfaces, wherein nongeometric factors such as bonding considerations specific to the emerging phase (or matrix) may play an important role in solid-solid transformations. Faceting of the new phase along an incoherent boundary segment during the nucleation stage can lower the nucleation barrier, ΔG^*, without bicrystallographic matching of the parent and product phases but rather because of energetic factors specific to the emerging phase. Subsequent growth of the new phase also may be essentially idiomorphic, exhibiting distinct facets on various length scales with such morphological features deriving primarily from the surface energetics and kinetic processes of atomic attachment characteristic of the crystallography of the growing phase. This notion of idiomorphic behavior during nucleation and growth has been promulgated by Soffa and co-workers[20,27] in their studies of the massive transformation in Mn-Al-(C) alloys and is inherent in the original work of Massalski and associates[15] on the massive transformation for many years. It is interesting to note that the formation of idiomorphic particles of solid Xe in supersaturated Al-Xe solutions[28] may be the inverse of the idiomorphic behavior described previously, in that the faceting may be dictated by the surface energy and kinetics of the surrounding Al-rich matrix. The facets appear to be identical to the faceting of voids in aluminum.[29] The idea that bicrystallographic matching or commensuration dominates virtually all interphase interfaces during solid-solid transformations is clearly a misconception, and incoherent/irrational interfaces in the absence of crystallographic constraints can play an important role in governing the morphology of the transformation products during nucleation and growth. Apparently, there are what might be called idiomatically "I-don't-give-a-damn interfaces"! (refer to the General Discussion of this 2004 Hume-Rothery Symposium). Of course, the importance of relatively disordered or incoherent interfaces in initiating and propagating solid-solid transformations goes back to Professor Cyril Stanley Smith's classic lectures of the late 1940s and early 1950s. The recent work of Vasudevan and co-workers[22,25] and Wittig[23] on Ti-Al-X alloys, Li *et al*. on Zr:ZrN interfaces,[26] and that of Soffa and co-workers on the Mn-Al-(C) alloys[20,27] has served to catalyze a reassessment of the nature and behavior of incoherent interfaces in solid-solid transformations. Also, the classification of interfaces by Howe *et al*.,[10] in which the type-3 incoherent interface was specified, made a very important contribution to this revival.

In the recent discussions of the nature of interphase interfaces and the role of bicrystallographic relationships between conjugate phases, some important new ideas have arisen that merit further attention both experimentally and theoretically. In particular, the matching of plane edges and atomic rows producing essentially collinear arrays or matching atoms between phases conjugated along type-3 incoherent interfaces or irrational habit planes appears to be an important fundamental feature of some interphase interfaces.[5–9] This 1-D coherency is thought to facilitate bonding across the interphase boundary and produce a reduction of the interfacial free energy. However, although some experimental results now exist supporting the role of 1-D commensuration in the evolution of microstructure in solid-solid transformations, the general influence of the shallow energy wells in the orientation space has yet to be established. In particular, the behavior of one-dimensionally commensurate interphase interfaces in the orientation space is expected to depend markedly on the depth of these wells relative to the effective driving force during interface migration. However, the position taken here is that during nucleation, the system will use any degree of freedom to effect a reduction in the interfacial free energy, thereby reducing ΔG^*.

Returning to the massive transformation, it is clear that nucleation energetics do not preclude a role for irrational, incoherent interfaces in the initiation and propagation of the product phase nor in the morphological evolution of the transformation front. The classic view of the massive transformation involving the migration of essentially incoherent interfaces through thermally-activated atomic jumping across this relatively disordered interface appears to be generally accurate, although the fundamental phenomena (continuous growth, ledge growth, faceting, *etc*.) may be influenced by such factors as one-dimensional commensuration of phases or idiomorphic behavior of the growing phase.

V. CONCLUSIONS

1. Incoherent interphase interfaces can be involved in the initiation (nucleation) and propagation (growth) of polymorphic and massive transformations as well as in various precipitation phenomena occurring in the solid state.
2. A relatively low energy incoherent interphase boundary can arise solely as a result of preferential formation of crystal faces associated with the natural habit of the emerging phase; that is, faceting on various length scales can be the result of thermodynamic, kinetic, and crystallographic factors primarily specific to the growing phase. This idiomorphic behavior can be virtually independent of geometrical matching and bicrystallographic factors across the interphase boundary.
3. Idiomorphic behavior can be a factor in both the nucleation and growth of intergranular and intragranular phase formation, as has already been noted earlier in connection with massive transformations.
4. So-called type-3 incoherent boundaries are indicative of the idiomorphic behavior of emerging phases.
5. Apparent incoherent planar boundary segments may also arise during the nucleation and growth of intergranular and intragranular phases as a result of plane edge-to-edge matching or 1-D commensuration of conjugate phases. The importance of this bicrystallographic matching or conjugation of phases is likely to depend on the depth of the energy wells in orientation space associated with these special orientations.
6. Both idiomorphic and bicrystallographic factors can influence the nucleation and growth of phases and their resultant morphologies in solid-state transformations.
7. The migration of incoherent interfaces can involve so-called continuous quasi-random jumping of atoms from the parent to product phase or a ledge mechanism depending on the nature and length scale of the boundary structure and driving force for the transformation.
8. Migrating incoherent interphase boundaries can be smoothly curved or exhibit facets on various length scales.

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