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On the massive transformation in γ-based titanium aluminides

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γ-based titanium aluminides are candidate materials for several high temperature structural applications. The orientation relationship, substructure and the interfacial structure of the massive product: matrix interface has been examined by transmission electron microscopy (TEM). The interfacial structure consisted of faceted, macroscopically planar interfaces, which were found to consist of regular arrays of ledges, no interfacial dislocations were observed. The substructure revealed a high density of planar defects such as twins, stacking faults and antiphase domain boundaries, such defects were also found in the vicinity of the interface. In another alloy, which had very few defects within the massive product, the expected orientation relationship between the product and parent phases was established. The low value of ledge height to interplanar spacing can explain the fast growth kinetics. The implications of these results on the understanding of massive transformations are discussed.

1. Introduction

The massive transformation is a diffusional phase transformation from one crystal structure to another, involving a rapid transfer of atoms across the interphase boundary, with no change in the overall composition. Massive transformations can occur during the decomposition of the disordered b.c.c. phases in alloys of the noble metals. The crystal structure change in such cases is mostly from the b.c.c. to f.c.c. phases or from b.c.c. to h.c.p. phases [1–5]. Such transformations also occur in other alloy systems, particularly in ferrous and titanium base alloys. Examples are the Fe-C, Cu-38 a/o Zn, Cu-20 a/o Al, Fe-Ni, Ag-Al, Cu-Ga, Ti-Au, Ti-Ag, TiAl based two phase alloys and other alloy systems. These transformations exhibit nucleation and growth characteristics, are thermally activated and occur during heating and cooling. Often, most of the atomic mobility is limited to the interphase boundary area. The term massive refers to the bulky or patchy morphology of the product phase, which is typically formed with fast growth kinetics.

In early studies, the *substructure* of the massive product was examined in several alloy systems [1–5]. In iron, massive ferrite exhibited simple dislocation patterns similar to that in equilibrium equiaxed ferrite, but differed a lot from the fine twinned or faulted structure of martensite. In Cu-Zn, the massive grains contained many large twins and very fine microwtins. The microtwins occurred at changes in the direction of faceted boundaries and at steps in the macrotwins. There were few stacking faults. The fact that few dislocations were associated with the massive/matrix interface suggested that (a) the massive growth is not associated with a dislocation mechanism and (b) that the transformation strains were not restricted to the developing massive/matrix interface. No regular dislocation arrays were observed.

In the Cu-Ga alloy system, the massive transformation resulted in a duplex product and exhibited a feathery structure. The internal structure of the massive phase showed complexities and the f.c.c. and h.c.p. phases were present together in lamellar form. The Cu-Ga system also showed twins, subgrain boundaries and dislocation networks within the massive product.

The common view on the *interfacial structure* at the massive: interfacial boundary is that the interfaces are usually incoherent and migrate by continuous growth in a manner similar to a high angle grain boundary. In some cases, however, growth can take place by the lateral movement of ledges across faceted interfaces [6]. According to Massalski, the growth of the massive phase proceeds by the migration of an interface that has either (a) the smooth curvature of a high energy interphase boundary or (b) faceted sections, which are often associated with a ledge mechanism. Massalski has provided examples of both situations [1–5]. The view of Aaronson and associates is that many of these boundaries are partially coherent [7–10]. Mou and Aaronson have shown that interfaces in the massive transformation in Ag-26 a/o Al were partially coherent and that the defects were misfit compensating or structural ledges [10].

The h.c.p. : f.c.c. type of transformation can serve as a model system to resolve this issue, since the kinetics and interfacial structure pertaining to growth are strongly governed by the crystallography of the two phases. An example of this type is the α to γ transformation in γ-based two phase Ti-Al alloys, where the h.c.p. (α) phase transforms to a tetragonal ($L1_0$ type, $a = 0.4$ nm, $c = 0.408$ nm) γ phase. At room temperature, the alloy system consists of two phases, α_2 and γ, the α_2 phase resulting from the ordering of the α phase. On cooling from the α phase field, a variety of morphologies can develop depending on the composition, cooling rate, holding time and other heat treatment variables [11–22]. For a given alloy composition, furnace cooling produces a lamellar structure, consisting of alternating plates of γ, twinned γ and the α_2 phases. Air cooling leads to a complex feathery and Widmanstatten morphology, while water quenching can produce a massive or martensitic transformation. The substructure of the product γ phase produced by the massive transformation (denoted γ_m, the subscript refers to the massive morphology) has received attention, since it offers insight into the phase transformation mechanism.

The interfacial structure between the parent α and the product γ_m phase has also been studied in TiAl based alloys, since the interfacial structure has a direct bearing on the growth kinetics and anisotropy of growth. However, contradictory results have been reported. According to Aaronson and coworkers, full coherency is general during the massive transformation and experimental evidence from the massive transformation in Ti-46.5 Al alloy was present [23]. The massive transformation was also investigated in Ti-47 Al and Ti-46.5 Al alloys [24, 25]. Zhang *et al.* have however, concluded that the interfacial structure is incoherent [26, 27]. To clarify these issues, the interfacial structure and substructure of the massive product of γ-based TiAl alloys was studied.

2. Experimental

An alloy of composition Ti-48Al-2Mn-2Nb was prepared by arc melting. Samples were then heat treated at 1420°C for 1 h, followed by water quenching. Thin foils were prepared for TEM studies from these samples. Another alloy of composition Ti-47Al-2Mn was prepared by arc melting, solutionized at 1390°C for 30 min. followed by water quenching. Thin foils of the heat treated samples were prepared for TEM studies from 200 μm thick, 3 mm diameter disks prepared in a twin jet polisher using a 65% methanol, 32% butan-1-ol and 3% perchloric acid solution. Immediately following perforation, the foils were gently rinsed in baths of methanol and distilled water. The TEM observations were conducted on JEOL 200 kV 2000 EX and JEOL 300 kV 3010 microscopes.

3. Results

TEM examination of the Ti-48Al-2Mn-2Nb alloy revealed (Fig. 1) a massive product (region (b) surrounded on one side, by the γ phase (region a) and on the other side by a two phase region α_2 and γ (region c) (Fig. 1). The massive product showed only a few defects, while the surrounding regions revealed a high density of planar defects. The orientation relationship (O. R.) between γ_m and the α_2 phase in region c was established as $\{111\}_\gamma // \{0001\}\alpha_2$ and $\langle 110\rangle\gamma // \langle 1120\rangle\alpha_2$.

TEM examination of the Ti-47Al-2Mn alloy revealed a massive product with an interfacial structure, which was faceted, and Fig. 2a shows a low magnification view of the massive region in the vicinity of the interface. The substructure was seen to consist of many planar defects, such as twins, stacking faults and anti phase domain boundaries (APDB). The twins were found by trace analysis to lie on {111} planes. The planar defects were visible within the massive region (Fig. 2b), as well as at the interface between the massive region and the α_2 grain (Fig. 2c). Note the faceted nature of the interface. The various planar defects interacted with each other, an interesting example being the intersection of a stacking fault with an APDB to produce a region having the same contrast as the perfect grain (Fig. 3).

As expected, the interfacial structure was found to vary with interfacial orientation. The presence of ledges at the interface was confirmed and a cross grid of ledges of average spacing 40 nm was observed (Fig. 4a and b). Importantly, such a cross grid of ledges, perpendicular to each other, was seen to completely bound the massive region in various orientations (Fig. 4c). Note that the variation of interfacial structure with orientation is particularly clear in Fig. 4c. In several orientations, a high density of planar faults was observed on the γ_m side of the $\alpha_2 : \gamma_m$ interface (Fig. 5a and b). The macroscopic habit plane was found by trace analysis to lie along the {111} plane.

At some orientations, a regular set of superledges was observed with a spacing of about 100 nm and height of 50 nm (Fig. 6a and b). The facets forming the terrace and riser of the superledges were found to be of the {111} and {110} type. Tilting experiments showed that the ledge height is approximately 3 nm (Fig. 7). A linear set of ledges was observed at orientations with a spacing of about 5 nm (Fig. 8). Another example from a different interfacial orientation can be seen in Fig. 9a. Each of the facets was found to consist of regularly spaced ledges of approximately 7 nm spacing (Fig. 9b). In the case of both planar and faceted interfaces, no misfit dislocations were observed.

4. Discussion

The massive transformation in TiAl based two phase alloys has attracted considerable attention. It is useful to study the massive product in this system since it is expected that crystallographic effects will be clearly brought out in a h.c.p. : f.c.c. type transformation. The role of composition and cooling rate in the formation of the massive product has been studied and it was shown that the massive product occurred, on water quenching from the α phase field, in alloys with a typical composition being Ti-46.54Al [19–22]. The present results show that the massive transformation also occurred on water quenching of Ti-47Al-2Mn and Ti-48Al-2Mn-2Nb alloys.

4.1. Substructure and orientation relationship

The present results showed that, in one of the alloys studied, a definite orientation relationship could be established between the massive product and the

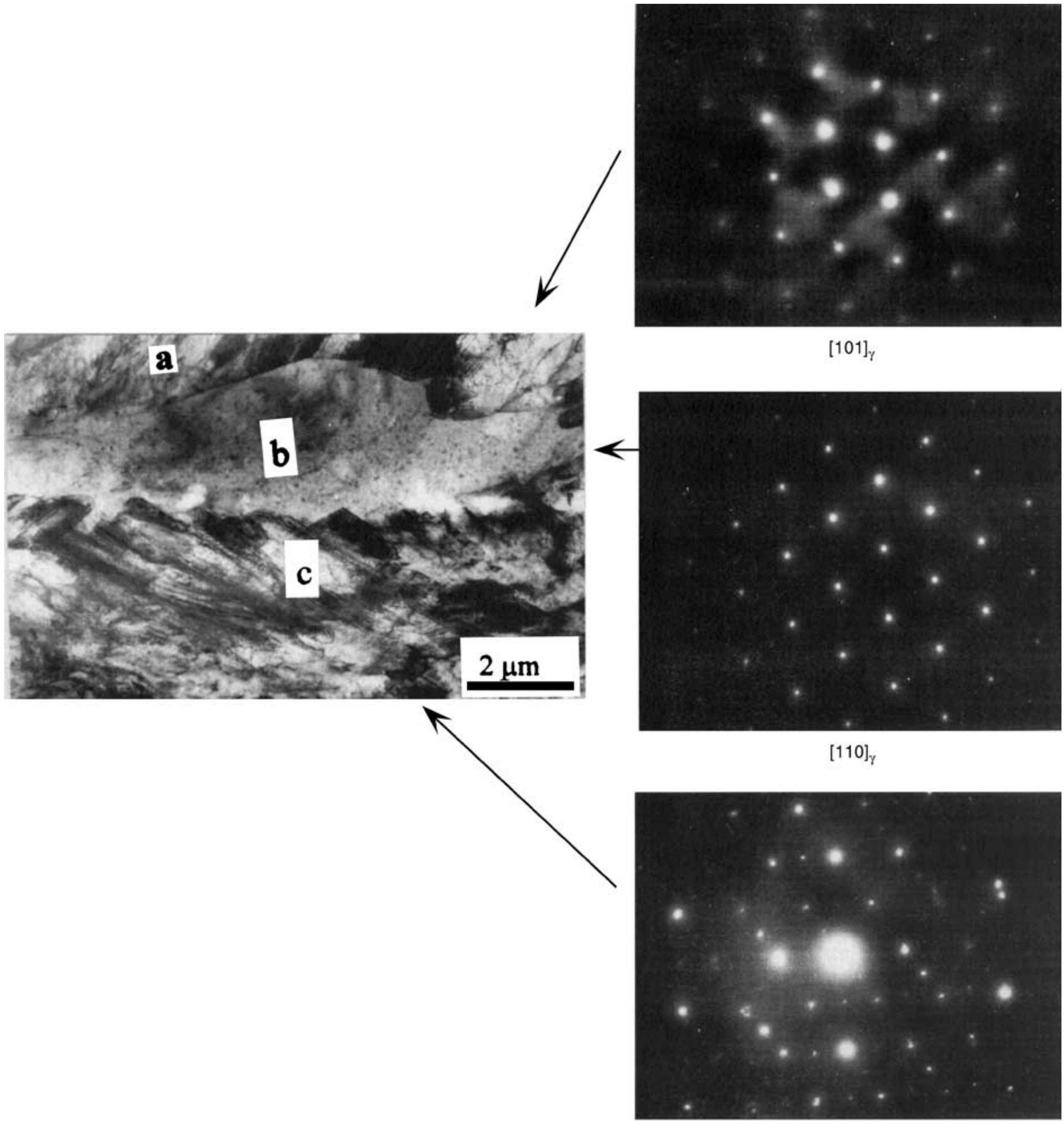
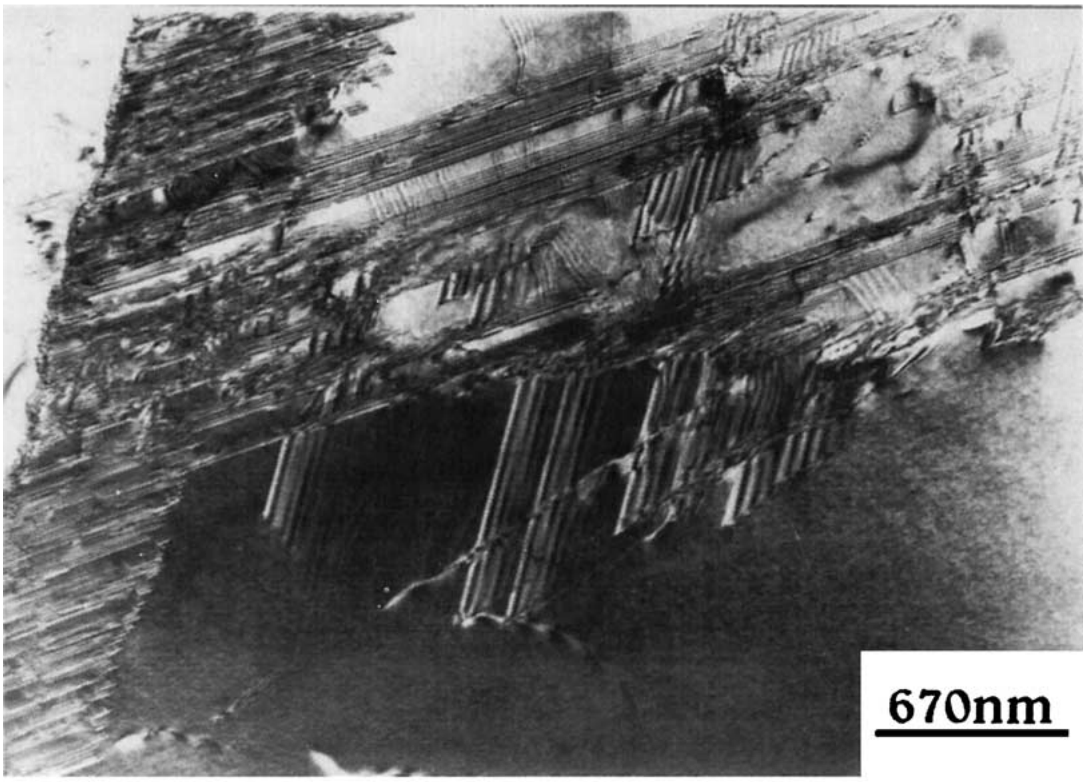


Figure 1 BF TEM micrograph of the massive region (region b) in a Ti-48Al-2Mn-2Nb alloy heat treated to 1420°C for 1 h. followed by water quenching. The massive region is surrounded by a twin related γ phase (region a) and a two phase $\gamma + \alpha_2$ region (region c). The O. R. between the massive γ phase and the α_2 phase in region c is $\{111\}_\gamma // \{0001\}\alpha_2$ and $\langle 1\text{-}10\rangle\gamma // \langle 11\text{-}20\rangle\alpha_2$.

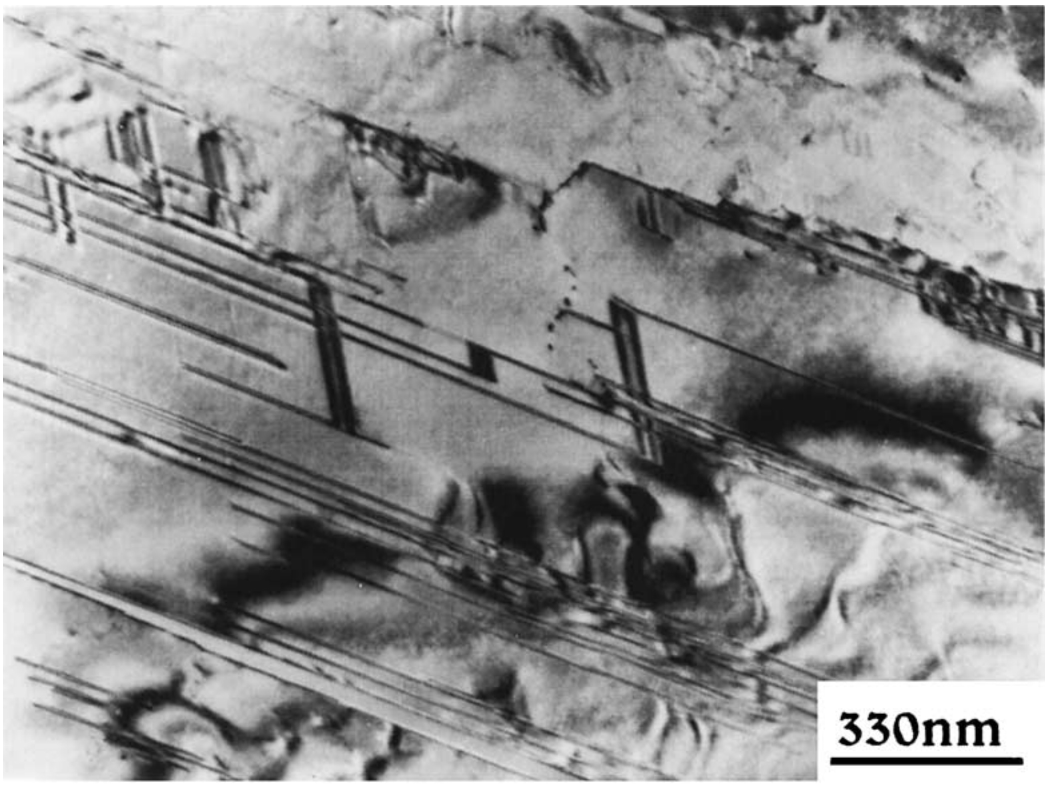
parent phase (Fig. 1). The substructure observed in the present investigation consisted of planar defects, including twins, stacking faults, APDB (Figs 2 and 3). Earlier TEM investigations had also shown that complex antiphase domain boundaries (APDB) and microtwin boundaries [26, 27]. Li and Loretto [28, 29] in their study of Ti-48Al-xNb alloys also found a dense array of planar defects, i.e., antiphase boundaries, stacking faults and microtwins. The present observations are consistent with these results.

The presence of the APDB in the substructure can be attributed to the formation of a metastable disordered f.c.c. phase from the parent α phase. The f.c.c. phase subsequently orders to the γ phase, resulting in the formation of APDB. The coupling of the defects, observed in the present investigation (Fig. 3), can be attributed to the reduction of energy by defect-defect interaction leading to a new defect with lower energy. The boundaries between antiphase regions were found in earlier investigations to be thin 90° domains (i.e., a domain where the c axis is rotated by 90° w.r.t. c axis of neighboring domains), separating adjacent domains with the same orientation that are yet out of phase [26, 27].

In their study of the massive transformation in γ-TiAl alloys, Denquin and Naka [30, 31] claimed that the nucleation of the γ_m phase took place mainly at the grain boundaries, through the formation of an ordered nucleus or a disordered f.c.c. nucleus, followed by ordering. Their TEM observations also showed a large



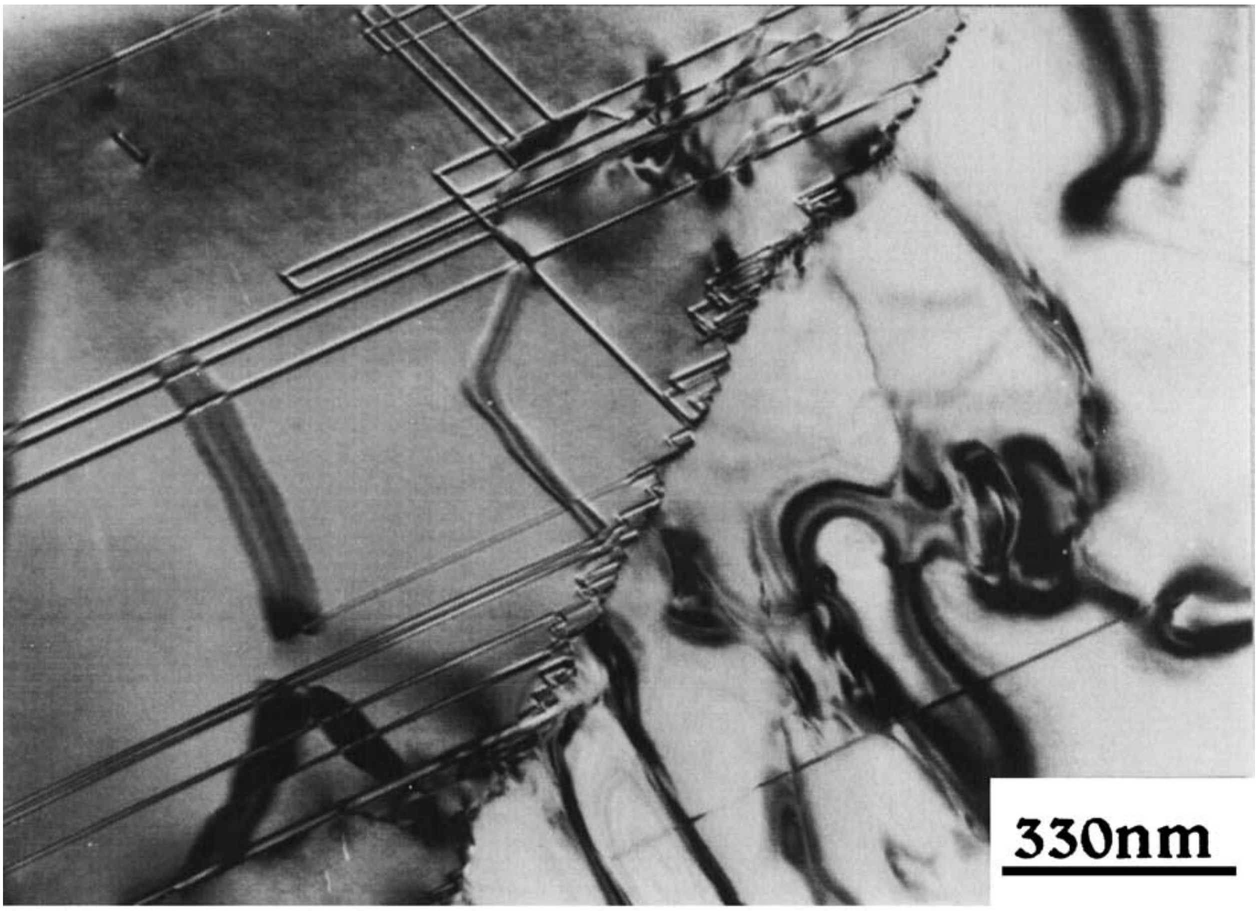
(a)



(b)

Figure 2 (a) BF TEM micrograph of a Ti-47Al-2Mn alloy, showing a massive region and the massive matrix interface. Note the high density of planar defects, twins, stacking faults and APDB and the high density of microtwins at the interface, (b) within the massive region, microtwins and stacking faults on {111} planes and the interaction between these defects, (c) at the massive : matrix interface showing microtwins and stacking faults on {111} planes. Note the faceted nature of the interface and the {111} and {110} type planes comprising the two sets of facets. *(Continued.)*

number of faulted domains, including stacking faults which lie on {111} planes and complex APDB. Two ordering processes, similar to the ones described above, were proposed to explain the substructure of the γ_m phase, (a) the α to γ transformation can result in the formation of order domains and antiphase domains and (b) if there is a metastable disordered phase, followed by ordering reaction at separate sites, the encounter of two growing domains can lead to the formation of order domains or APBs.



(c)

Figure 2 *(Continued.)*

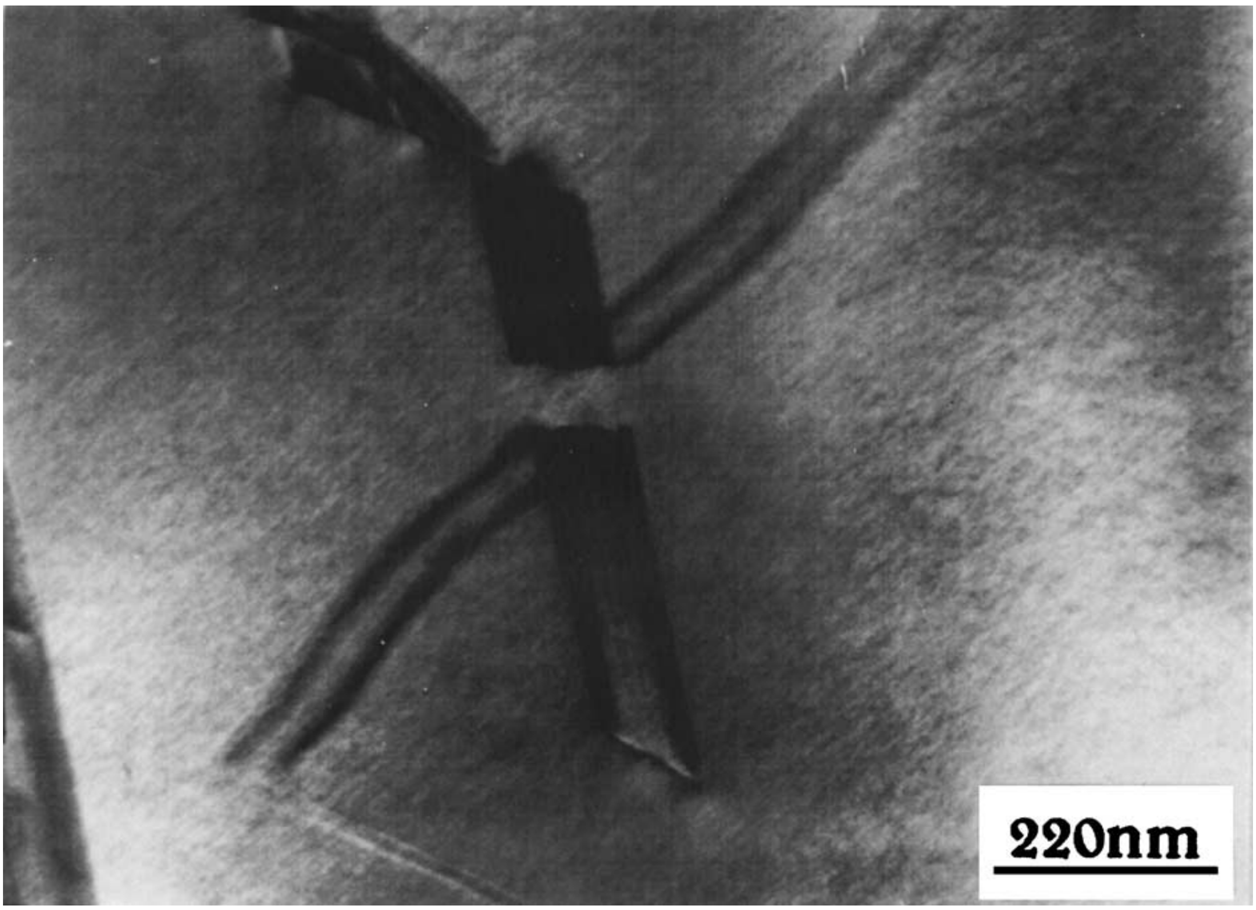
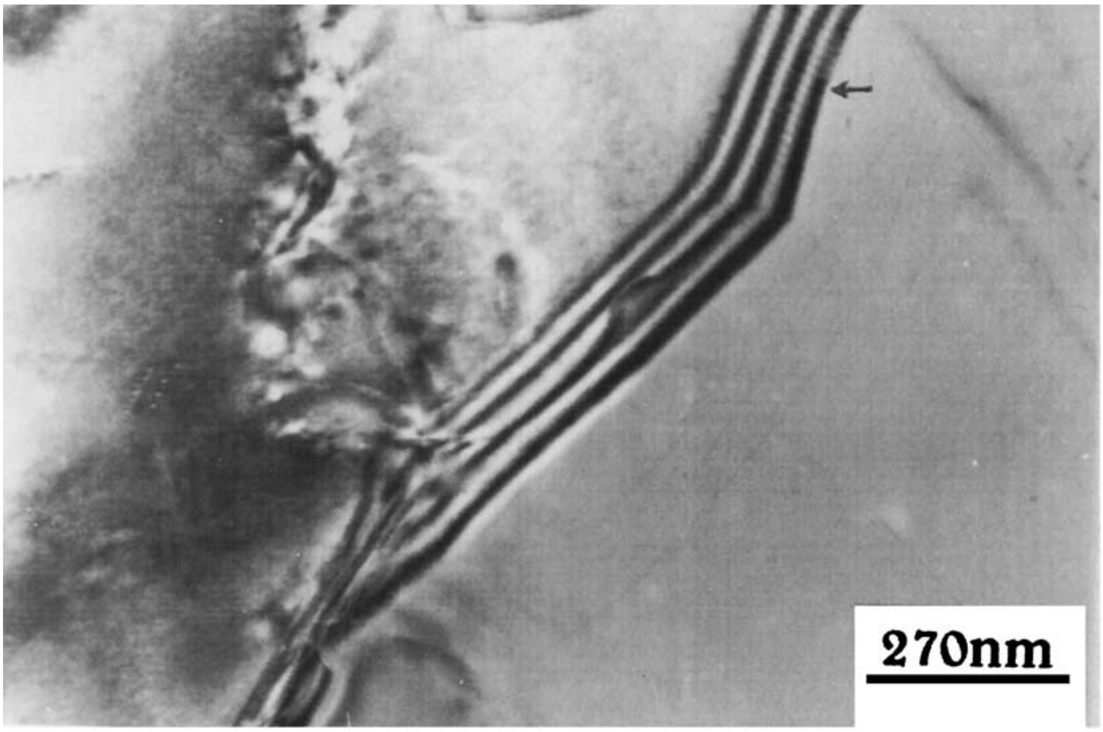
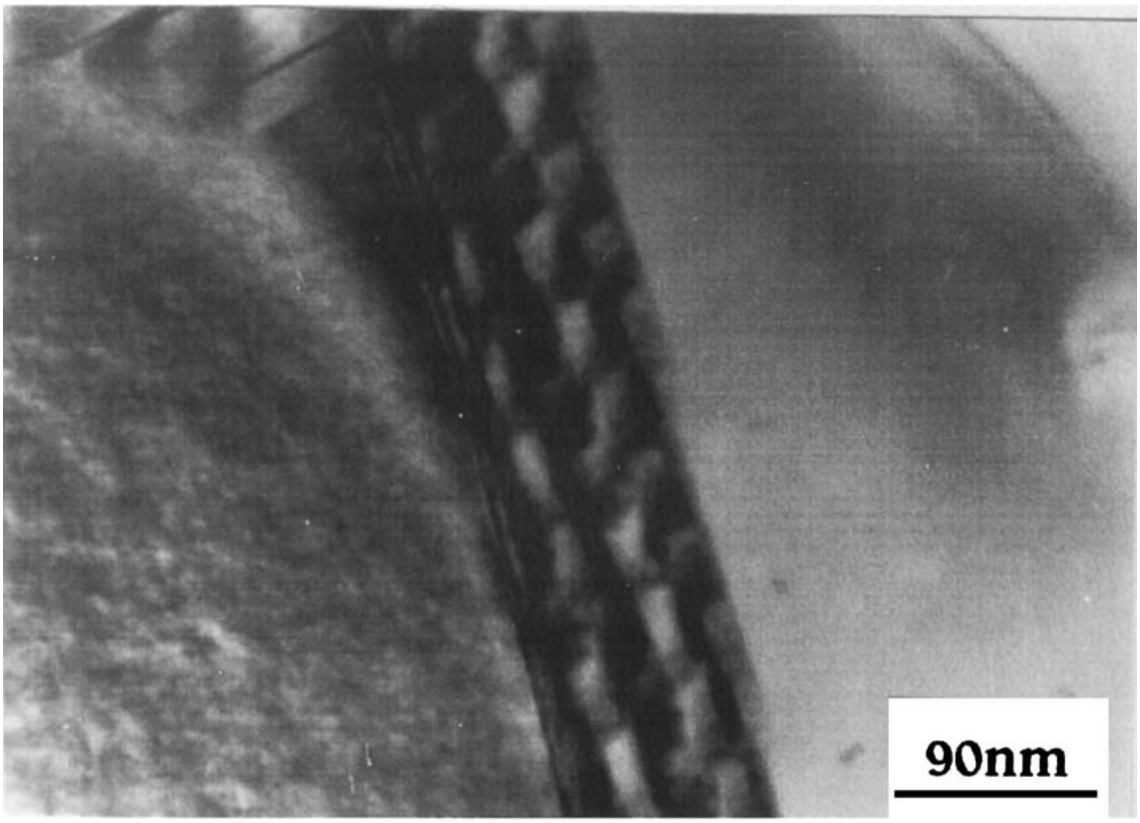


Figure 3 An example of the interaction between defects to produce a region having the same contrast as the matrix.



(a)



(b)

Figure 4 (a) Planar massive : matrix interfaces at various orientations. Note the ledge structure clearly visible on the upper interface, (b) Higher magnification micrograph of the planar interface, showing a perpendicular cross grid of ledges of spacing of about 40 nm, (c) massive : matrix interfaces at various orientations. Note the crystallographic nature of the interfaces, the cross grid of ledges and the pronounced faceting at the interfaces. *(Continued.)*

Observations and analysis of defects within the massive phase were performed [24]. These observations showed that the defects were (a) dislocations, (b) stacking faults and (c) antiphase boundaries associated with dislocations and stacking faults. The present results on the substructure are in conformity with the observations cited above. Planar defects such as APDB, stacking faults and microtwins were observed and several cases of defect-defect interaction were noted. A high density of twins was noted at the vicinity at the massive: matrix interface (Fig. 5), possibly resulting from the stress generated in the crystal structure change from the parent to

(c)

Figure 4 (Continued.)

the product phase, as well as from the thermal stresses generated during cooling from the solutionizing temperature.

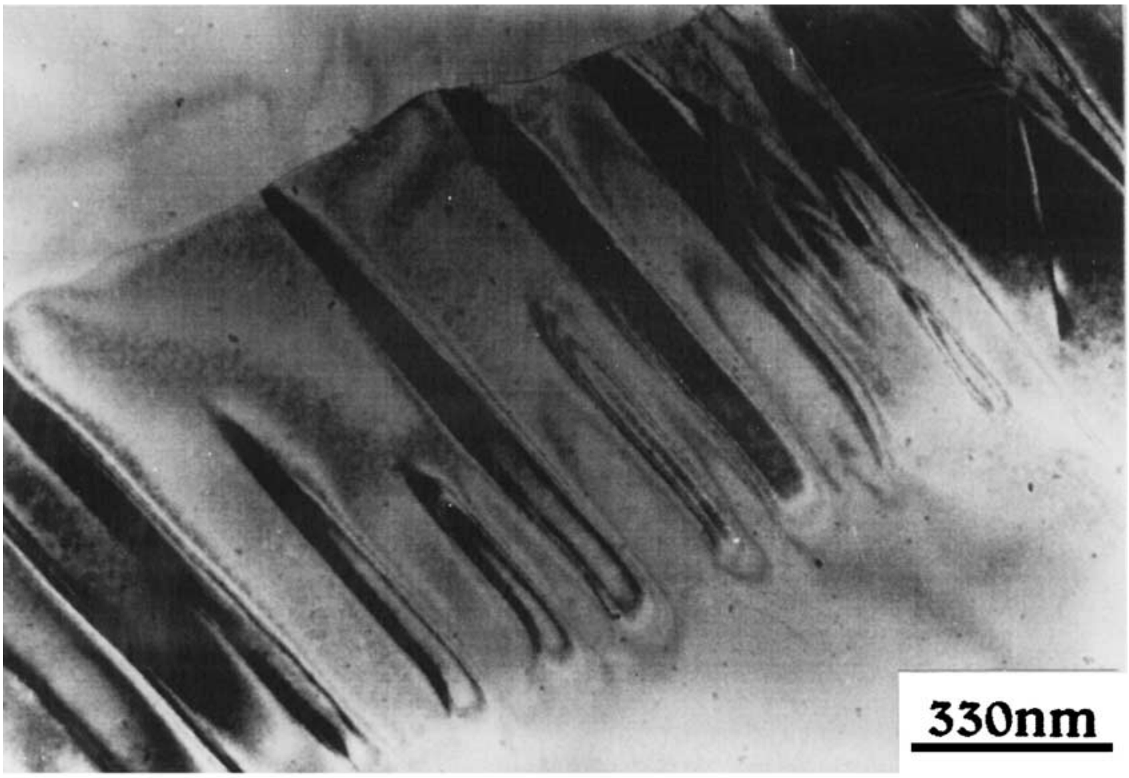
4.2. Interface structure

The structure of the interface between the massive product and the matrix will now be discussed. In $\gamma + \alpha_2$ alloys, *Denquin* and Naka claimed that the nucleus of the massive phase were coherent with one of the two grains [30, 31]. This coherency was assumed to hinder diffusion across the matrix: nucleus interface, leading to growth of γ_m into the opposite grain by diffusion across an *incoherent* α/γ interface. They found that the boundaries delimiting the regions, massive and lamellar were irregular and curved. No simple orientation relationship was observed between the regions. The occurrence of twinning could have affected evidence of O. R., such twinning could have been initiated by a transformation induced stress concentration.

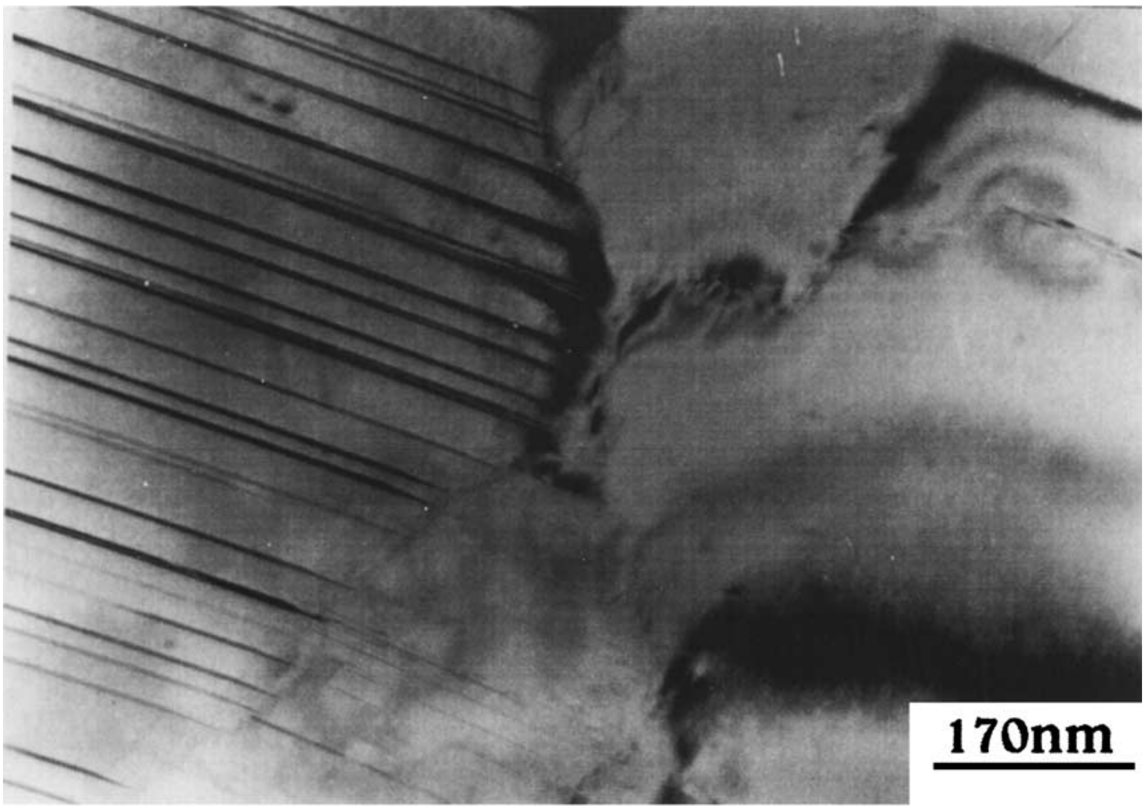
On the other hand, in a comparative study of the massive transformation in Ti-46.5Al and other alloy systems, *Aaronson et al.* found ledged, planar boundaries in Ti-46.5Al [23]. A rational O. R. was found to be lacking. Linear interfaces were present and superledges often appeared at these interfaces. Such interfaces were possibly parallel to {111} planes. A planar $\gamma_m : \alpha_2$ boundary was bound to be fully coherent except for a few stacking faults in the vicinity of the interface.

In another investigation by Veeraraghavan *et al.*, nucleation of the massive phase was found to occur at grain boundaries [25] and interfaces were found to have both curved parts as well as planar facets. Superledges were often seen on the planar facets and the facets were free of misfit dislocations. Higher magnification observations revealed a number of closely spaced defect structures, reminiscent of DSC dislocations along some facets and especially along the superledges. From this information and the interfacial structure, it was discussed whether growth took place by the ledge or the continuous growth mechanism.

According to Loretto and coworkers, the advancing γ_m interface tended to facet parallel either to one of the {111} planes or to the basal plane in the grain being consumed due to its impingement on the existing γ lamellae [26–29]. Thin microtwins and α_2 platelets then form in γ_m perhaps due to transformation stresses and supersaturation. No O.R. and no coherency between the γ_m and the α grain, which is consumed by the massive front, was found. The nature of the γ_m: matrix interface was studied and the view of Aaronson *et al.* was disputed. Instead, Loretto *et al.* claimed that there was no O.R. between the phases and no indication that the interface was coherent. This conclusion was based on the examination of a number of regions where the γ_m phase had grown into the adjacent α grain. The interface between γ_m and a lamellar region in a Ti-48Al alloy was clearly faceted, but there was no rational O.R. between the phases. The facets along the massive interface were approximately parallel to one of the {111} planes in γ_m, as noted by comparing the traces of the {111} features within the γ_m or the lamellae in the adjacent lamellar grain.



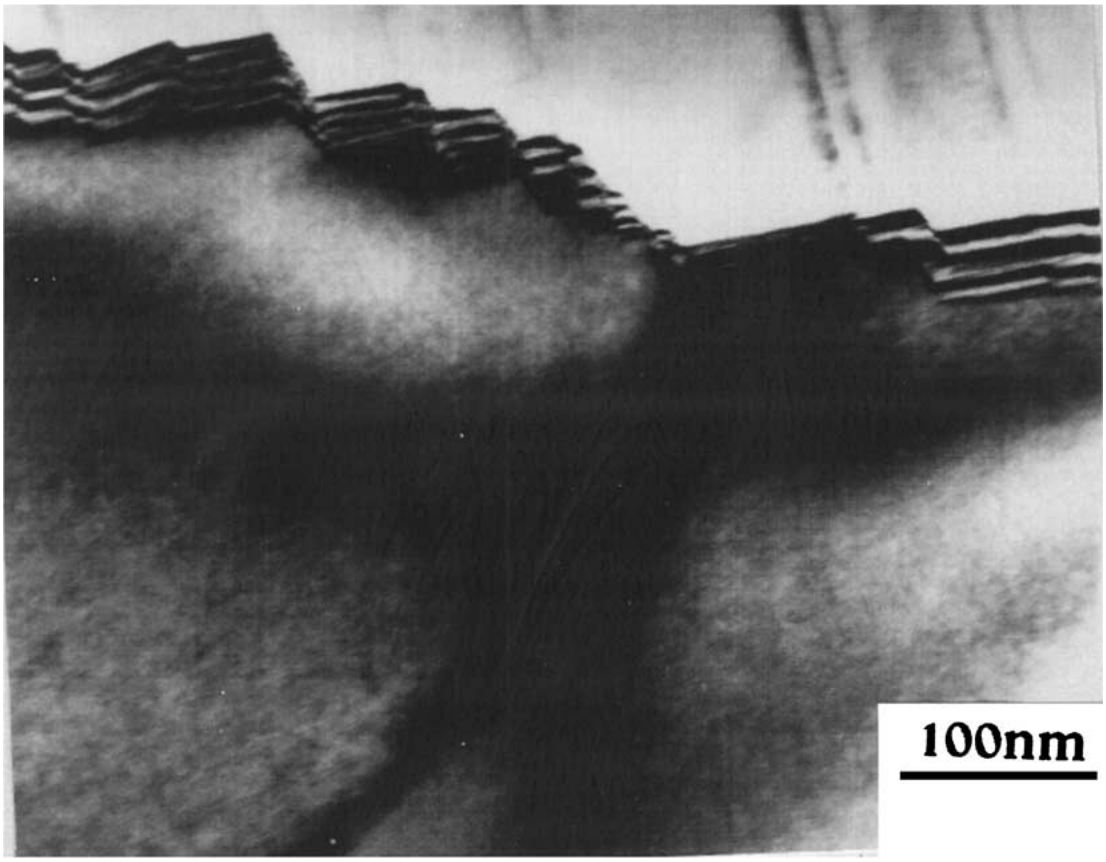
(a)



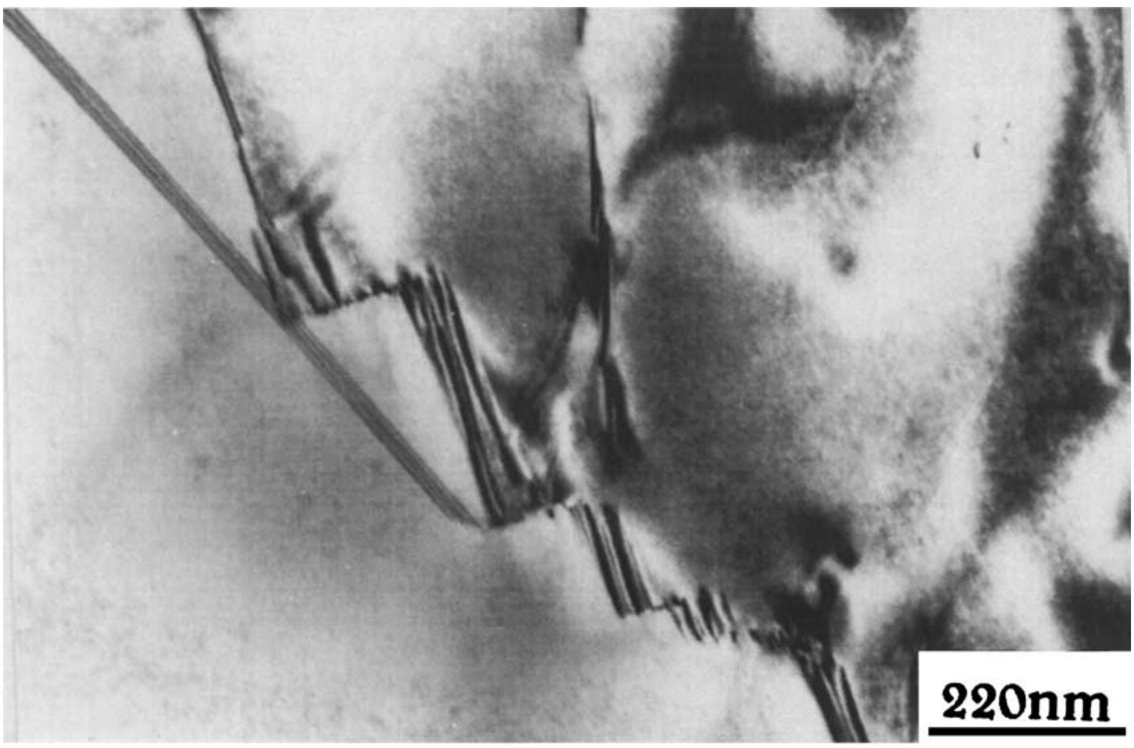
(b)

Figure 5 (a) Parallel planar defects at a massive : matrix interface with a spacing of about 200 nm, (b) another interfacial orientation with parallel twins of average spacing 80 nm.

Thus there is no unanimity in the interpretation of the nature of the interface between γ_m and the grain into which it is growing. However, the present results (Figs 4 and 6) clearly show numerous instances of faceted interfaces (Fig. 4c, for example), superledges (Fig. 6), a regular ledge structure (Figs 7–9) and the absence of Shockley partials at the interface. Clear evidence of faceting (Fig. 9) has been presented. The results contradict the view of Zhang *et al.* and Denquin and Naka that the interface is incoherent by providing examples of ledged structures. In addition, the faceted habit planes, observed in the present investigation, showed



(a)



(b)

Figure 6 (a) Superledges at the massive : matrix interface also showing a defect-interface interaction, (b) superledges with a spacing of about 100 nm and a ledge height of approx. 50 nm.

the importance of crystallography in the growth of these interfaces. Moreover, the establishment of the O.R. between γ_m and the α_2 phase as $\{111\}_\gamma // \{0001\}\alpha_2$ and $\langle 1\text{-}10\rangle_\gamma // \langle 11\text{-}20\rangle\alpha_2$ in the quaternary alloy showed that the conventional theory of nucleation and growth is applicable to the massive transformation in this alloy. The presence of twins and other defects in the massive phase could have obscured the O.R. in the ternary alloy.

The implication of the present results for the growth kinetics is that it is possible to reconcile the fast growth kinetics with the ledge structure. The growth rate of an interface containing ledges with height h and interledge

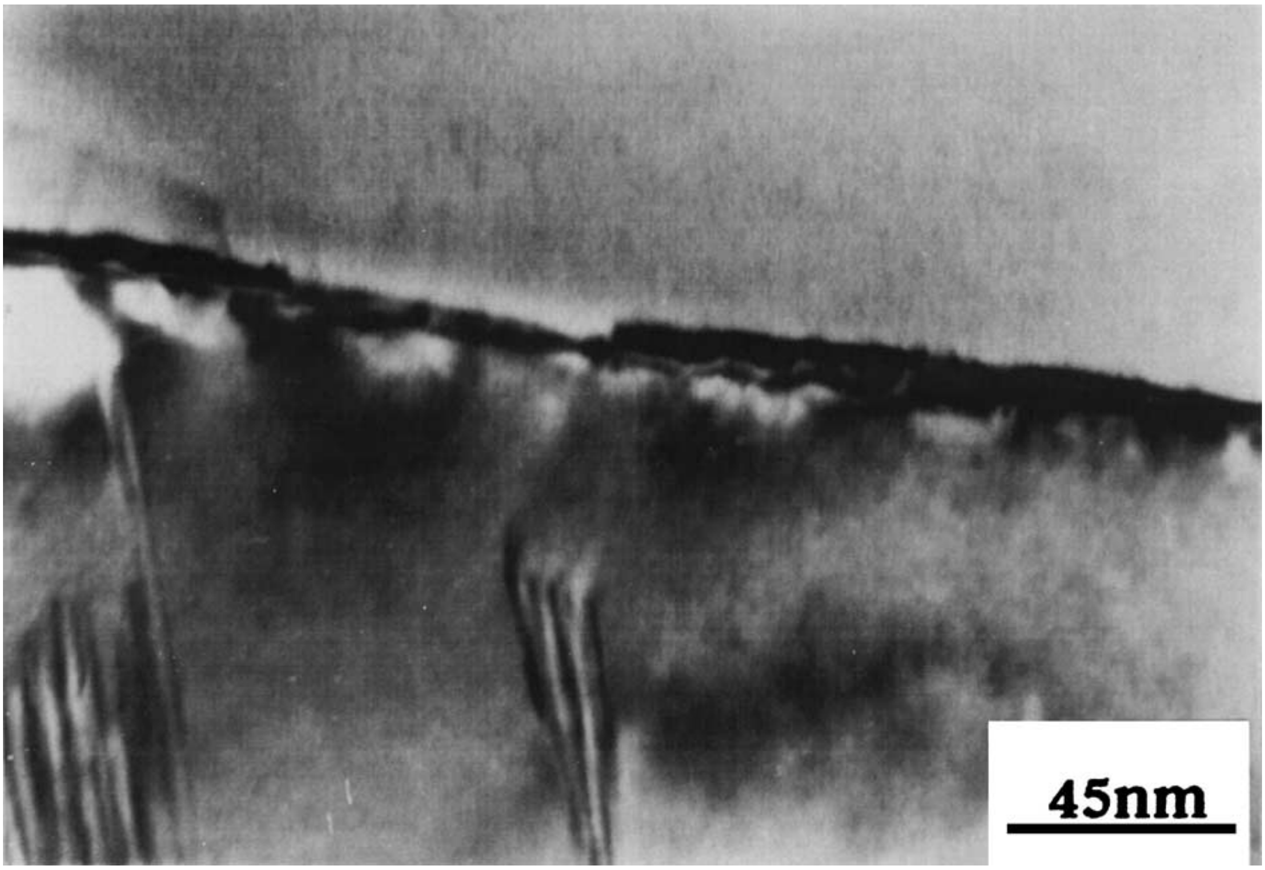


Figure 7 Ledge of height 3 nm found at the massive : matrix interface.

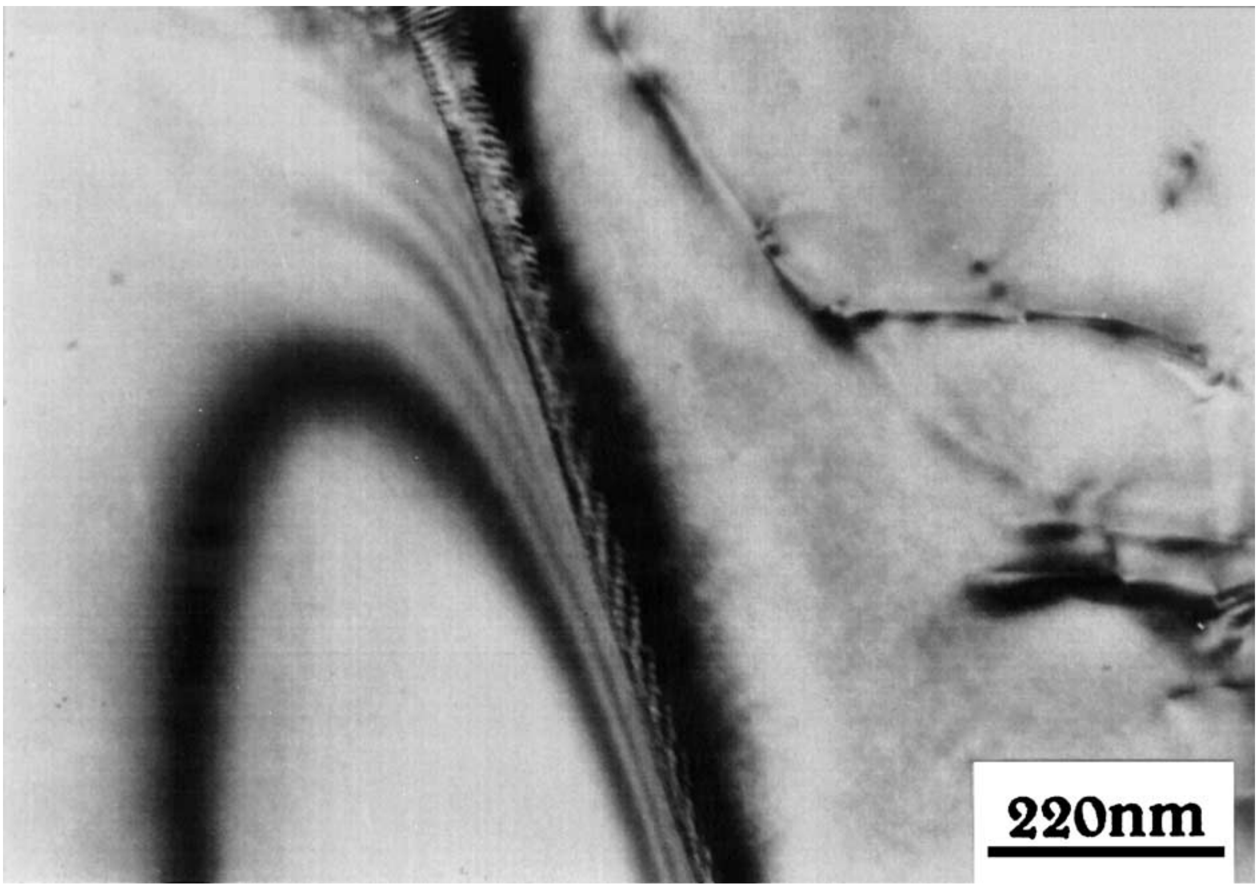
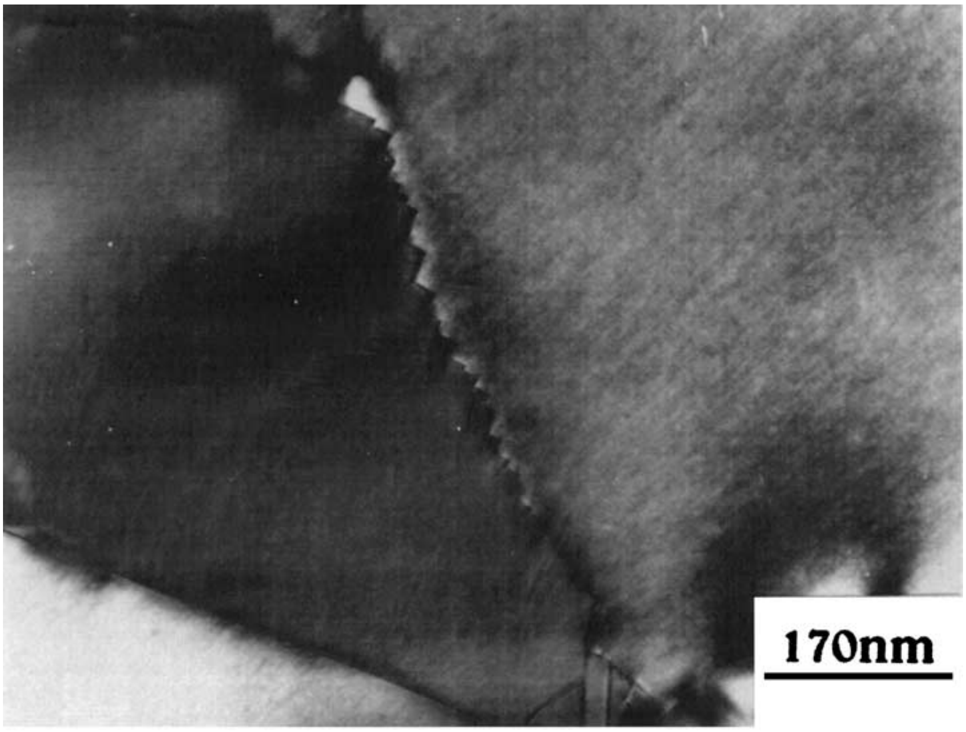
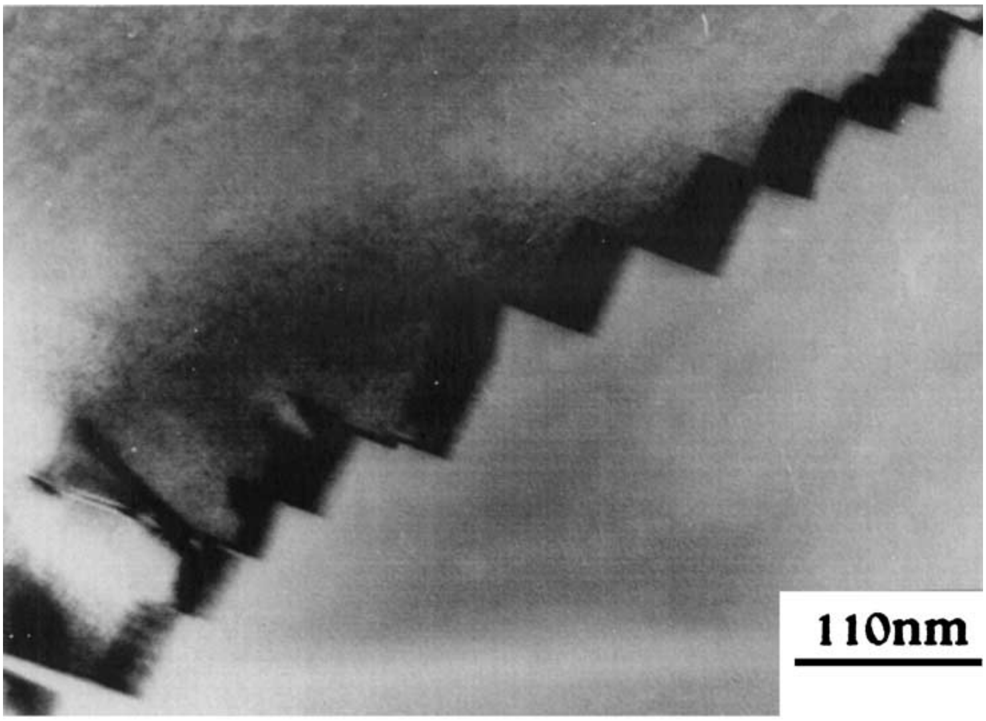


Figure 8 BF TEM micrograph of a massive : matrix interfacial orientation with a regular ledge structure.



(a)



(b)

Figure 9 (a) BF TEM micrograph of faceted and planar orientations of the massive : matrix interface, (b) Higher magnification micrograph of the faceted orientation of the massive : matrix interface. Note the ledge structure visible on the top and bottom facets.

spacing 1 is $h/1$ times that of an incoherent boundary. Our finding is that $1/h$ is in the range from 1 to 10, thus the fast growth associated with an incoherent boundary can be mimicked by the ledged interfaces. The superledges can form by consolidation of the ledges.

5. Conclusions

The orientation relationship, substructure and the interfacial structure of the massive phase in two phase $\gamma + \alpha_2$ alloys was studied by TEM. The massive product contained a large number of planar defects both within the massive grain and in one case, at the massive: matrix interface. The interfacial structure was found to be composed of either planar features with ledges or faceted planes perpendicular to each other. In both cases, regular sets of ledges were observed and no misfit dislocations were seen. The expected O.R., i.e., the parallelism of close packed planes and directions in the product and parent phases, was observed in one of the alloys studied. The low value of ledge height to interledge spacing makes the fast growth kinetics of the massive phase feasible.

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