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**Coherent joining of the domains in Mg<sub>2</sub>Al<sub>3</sub> β and β' phases**

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## Coherent joining of the domains in $Mg_2Al_3$ $\beta$ and $\beta'$ phases

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The structure of two phases, the  $\beta$  and  $\beta'$  of  $Mg_2Al_3$  metallic compound has been described in terms of rod-like domains consisting of periodic sets of hexagonal layers. The coherent joining of the neighbouring domains has been discussed. The main set of 11 layers forms a stable configuration which does not change during the  $\beta' \rightarrow \beta$  transformation. The short sequences of 2-3 layers presented on the  $\beta'$  phase transform into clusters emerging in the  $\beta$ -phase.

Keywords: complex metallic alloys; Mg-Al compound; Samson phase

### 1. Introduction

The  $\beta$ - $Mg_2Al_3$  Samson structure is an example of the complex intermetallic structure intensively investigated by Samson [1] and many other authors (for the review see [2]). The Samson phase is stable at a temperature above 214°C. Its elementary cell contains 1,168 atoms distributed over 1,832 atomic positions of the face centred cubic - FCC crystal lattice ( $Fd\bar{3}m$ , no. 227 space group,  $a_c=2.8242(1)$  nm). The structure's skeleton consists of 528 aluminium atoms and 351 magnesium atoms. The site occupation factor (SOF) is equal to one for all the skeleton atoms and they are arranged in a sequence of hexagonal layers [3,4]. The remaining atoms, with  $SOF < 1$ , partially occupy their structural positions and many of them belong to clusters arranged in a tetrahedral lattice as it has been described in detail in [1,5,6].

The  $Mg_2Al_3$  compound structure transform into one of the subgroup of the  $Fd\bar{3}m$  group - the space group  $R\bar{3}m$ , no. 160 below 214°C. The resulting rhombohedral  $\beta'$ -phase has the following lattice constants written in the hexagonal frame:  $a_r=1.9968(1)$  nm and  $c_r=4.89114(8)$  nm. One obtains the same experimental values of the lattice parameters for both phases by also using the hexagonal coordinates for the FCC structure. This means that there are only some local atomic position changes during the  $\beta \rightarrow \beta'$  transition.

It is well known that many elements crystallize in the close packed structures (*e.g.*: the hexagonal close packed structure – HCP or the face centred cubic structure – FCC). The two components of the discussed  $Mg_2Al_3$  compound, aluminium and magnesium, also belong to those elements. A pure aluminium crystal lattice is FCC. In the case of Mg atoms, they crystallize in the HCP lattice. The inter-atomic distance of the nearest neighbouring atoms is about 12% larger for magnesium than the corresponding one for the aluminium atoms. Both structures are close packed, although they consist of different sequences of hexagonal layers. The sequence is ABC for the cubic (FCC) structure and AB for hexagonal (HCP) structure. Such configurations of layers are presented in the close packing of hard spheres.

One should also remember that the ABC sequence of hexagonal layers is characteristic for all the cubic structures. It is present not only for FCC structure but also for other cubic lattices: simple cubic (SC) and body centred cubic (BCC). However, only every second atomic position in the main hexagonal plane is occupied for the last structures.

The structural features similar to the Samson structure have already been observed in some other complex phases, *e.g.*:  $\text{Cu}_4\text{Cd}_3$  [7] or  $\text{NaCd}_2$  [8], where the regions corresponding to the hexagonal domains consisted of fragments of the Laves phase structures. The Laves structures are frequently described in terms of the stacking of hexagonal layers and also by commensurate modulation [9].

## 2. Sequence of hexagonal layers

The hexagonal layer sequence in the  $\beta\text{-Mg}_2\text{Al}_3$  structure has been recently published [3,4]. The atomic positions obtained by Feuerbacher *et al.* [2] were used. All the length values were given in relation to the lattice constancies of the hexagonal reference frame.

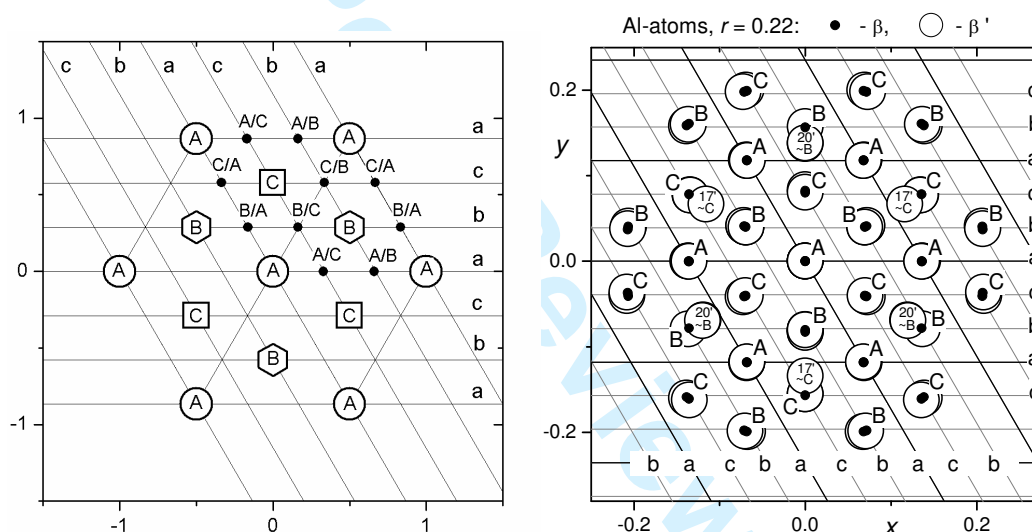


Fig.1. (*left*) Three types of stable atomic positions (A, B or C) for hexagonal lattice of close packed hard spheres. In the figure, intersection points of various lattices (a, b or c) are also presented – for instance A/B or A/C. Such intersections are denoted in the text by a letter M ('M' for Mixed). (*right*) The skeleton Al atoms of the inner part ( $r=0.22$ ) of single domain projected onto the (111) plane for the cubic structure  $\beta\text{-Mg}_2\text{Al}_3$  (black dots) and compared to the projection of all Al atoms onto the base plane of the rhombohedral  $\beta'$ -phase (open circles). All the Al atoms of  $\text{Mg}_2\text{Al}_3$  occupy A, B or C positions of the hexagonal lattice. Three atoms of layer 17' and three of layers 20' are shifted from the perfect hexagonal positions for the  $\beta'$  phase only (the position of particular layer is given in Figure 2).

Figure 1 (*left*) presents a hexagonal layer and positions that can be occupied by atoms within this layer. Lines denoted with symbols *a*, *b*, *c* determine the nodes of a hexagonal lattice. An intersection of two *a* lines leads to an A node. The same holds for lines *b* and *c* and the corresponding nodes B and C. Positions A, B and C correspond to the energetic minima of a rigid spheres set. In Figure 1 one can find other lines interceptions, as well. The resulting nodes are denoted as A/B, A/C and so on. In the further part of the paper symbol M (as 'Mixed') is assigned to all such

nodes. These are not points of energetic minima for rigid spheres sets, however, due to their symmetry; they form another commensurate structure.

In Figure 1 (right) Al skeleton atoms, that is all atoms for the  $\beta'$ -phase and 75% of atoms of the  $\beta$ -phase, are projected onto basal plain for single domain. Excluding six atoms of the  $\beta'$ -phase, within each domain all other atoms occupy commensurate positions of the A, B, C or M-type. These positions are occupied in both the  $\beta$  and  $\beta'$  phases. Six positions (open circles without a dot in their centers) are incommensurate with the other ones. This leads to a local instability and, in consequence to a clusters' creation. This type of clusters is characteristic of the  $\beta$ -phase.

There are rod-like domains along the main diagonal of the cubic unit cell arranged in the FCC lattice (Figure 2). The 22 layers (only half of them, namely 11, are the skeleton atoms) form the structural domain which consists of about five hundred atomic positions (many of them are only partially occupied).

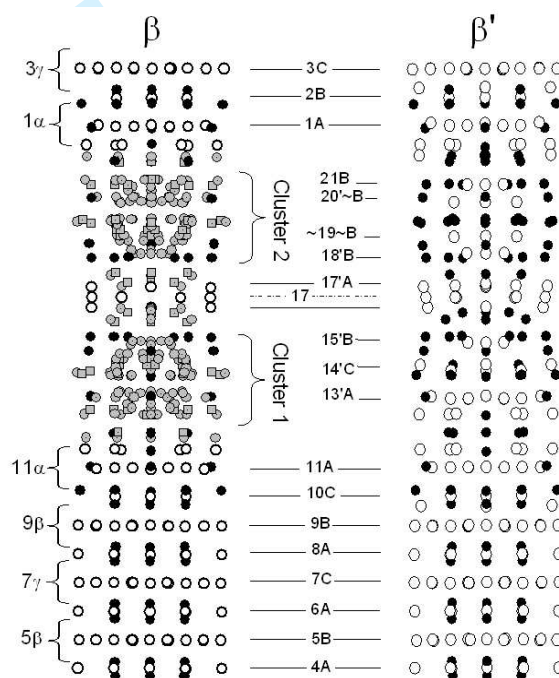


Figure 2. The single domain of the  $Mg_2Al_3$  intermetallic compound. The rod-like domain is oriented along the  $[111]$  direction for the cubic  $\beta$ -phase and the  $z$ -direction for the rhombohedral  $\beta'$ -phase. Open circles represent Al atoms, and full circles – Mg atoms. The black and white symbols describe atoms with  $SOF=1$ , grey symbols correspond to positions with  $SOF<1$ . There are 11 layers of Al atoms occupying A, B or C hexagonal positions for both phases. The Al layers with the neighbouring Mg layers form the  $\alpha$ ,  $\beta$  or  $\gamma$  slabs. The two cluster positions are marked for the  $\beta$ -phase. They correspond to the short sequences of hexagonal layers in the  $\beta'$  phase, namely three 13'-15' layers and two 17'-18' layers.

The main sequence of 11 hexagonal layers does not change during the transformation from  $\beta'$  to  $\beta$  phase for the  $Mg_2Al_3$  intermetallic compound. This part of the structure can be also constructed with the Friauf polyhedra discussed by Feuerbacher *et al.* (2007). If the entire space is filled by Friauf polyhedra, the ratio of Mg atoms to Al atoms would assume a 1:2 value instead of the real 2:3 ratio. The Mg atoms would be missing and the chemical balance is restored by Mg atoms scattered

within the outer parts of the structural domains, where the structure of  $\beta$  and  $\beta'$  phases are significantly different.

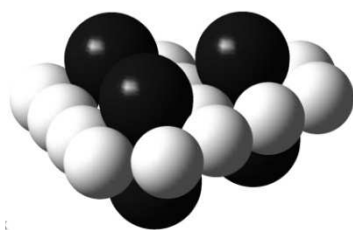


Figure 3. The central part of  $3\gamma$  slab of the  $Mg_2Al_3$  structure. Al atoms hexagonal rings with Mg atoms placed symmetrically in the centres of the hexagons. All the atoms are placed in C positions of the hexagonal lattice. For  $\alpha$  or  $\beta$  complexes the atoms occupy the A or B positions respectively.

At the end of the main sequence of 11 layers, one can observe transitional areas where Al and Mg atoms swap places. The structure changes afterwards into two short chains of hexagonal layers (Figure 2): a three-layer chain (layers no. 13', 14' and 15') and a two-layer chain (layers no. 17' and 18'). The short chain keeps the inter-layer distances; however, they are shifted, as a whole, with respect to the 22 reference layers. Layer 17 is filled with Al atoms occupying mixed positions marked by the letter M. Such mixed positions are some commensurate positions between different A, B or C points of the hexagonal plane.

The clusters are formed in the  $\beta$ -phase outside the main chain of 11 hexagonal layers (Figures 2 and 4). They are conglomerates of atoms which partially occupy their atomic positions in most cases ( $SOF < 1$ ). The spaces between the well ordered hexagonal layers and the clusters are filled with Mg atoms which partially replace the outer Al atoms (this is the case of layers no. 11 and 1). Conversely, within the neighbouring Mg layers, Al atoms partially replace the outer Mg atoms. This replacing mechanism leads from 11 ordered hexagonal layers to some crystalline disorder and a formation of the atomic clusters for the  $\beta$ -phase.

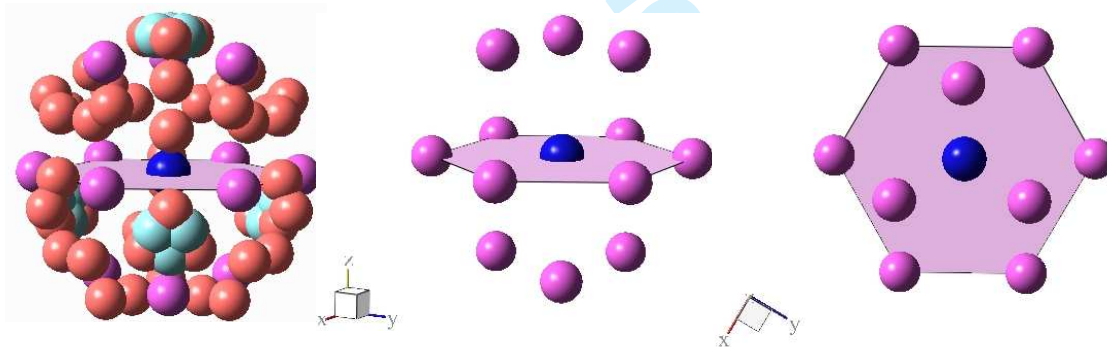


Figure 4. The local arrangement of atoms for the  $Mg_2Al_3$   $\beta$ -phase clusters. A magnesium atom (dark ball) is placed in the centre of the hexagonal ring of aluminium atoms (light balls).

The  $Mg_2Al_3$  compound structural stem is a set of stacked hexagonal layers filled interchangeably by Mg and Al atoms. In the case of the  $\beta'$ -phase (the lower-temperature phase) these layers are arranged into shorter and longer sequences of hexagonal layers and a transition from one sequence to another occurs by exchanging one type of atoms for another: Al for Mg and Mg for Al. This results in a very large elementary cell.

### 3. Coherent connection of domains

Figure 5 schematically shows a sequence of layers for the three neighbouring domains of the  $\beta'$ -phase (the I single domain is shown in Figure 1). The arrangement of layers within each domain is identical. The entire set consists of 22 parallel and equally-spaced layers. The distance between the two layers equals  $1/22 \approx 0.0788$ . The figures show only atoms which lie inside the domains: Al atoms – the thick lines and open circles; Mg atoms – the thin lines and full circles. The relative shift of the neighbouring domains along the  $z$ -axis is equal to  $1/3$ . In the figure, three different regions located around  $z$  equal to 0.25, 0.383 and 0.647 are shaded. We used these regions for an analysis of the coherency between the different domains.

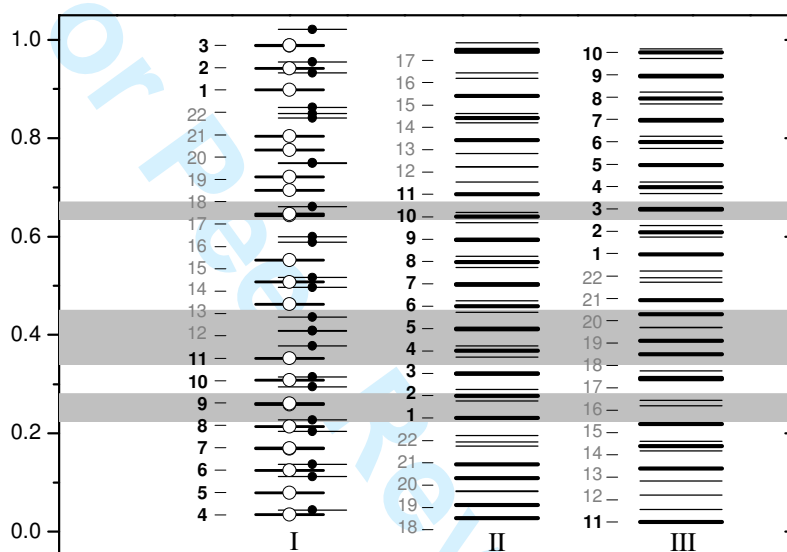


Figure 5. The sequence of 22 hexagonal layers for the  $\beta'$  phase and their arrangement within the particular domains: I, II and III. The thick lines indicate Al layers, thin ones – Mg layers. All three domains are identical. They are shifted by  $1/3$  with respect to each other along the  $z$ -axis. The structure of the inner area of the first domain is outlined in the figure in detail. The open circles are associated with Al atoms and full circles with Mg atoms. The cross-sections indicated by three shaded regions are analyzed in detail in the text.

Figure 6 outlines a cross-section of the  $\beta'$ -phase at the level of  $z=0.25$ . The areas of particular domains are marked with continuous lines. Domain I (with layers 2B of Al and Mg) and domain II (with layers 1A and 2B) are significantly enlarged. Their size considerably exceeds the area of the circles which determines the maximum size of the close-packed hexagonal domains. Domain III, on the other hand, although its border is marked in the figure, is completely devoid of atoms. Such an observation leads to a conclusion that for this particular cross-section, domain III vanishes while domains I and II extend by taking over the area of domain III. It should be also emphasized that domains I and II border on each other in a coherent manner. For this cross-section, there are no atoms that would not belong to either of those two domains (this holds true for all Al and Mg atoms).

In Figure 7, one can find a cross-section (its  $z$  coordinate is equal to 0.647) within which all three domains are filled with atoms in a fairly uniform manner. This cross-section is an equivalent to  $\beta'$ -phase's layer 17'. The conclusion that follows

from the figure is that all three domains cannot merge in a coherent way. Domains II and III are, indeed, connected in such a way and at their edges, there are atoms of M type coming from domain I. Conversely, the edge of domain I is occupied by Al atoms being placed at  $\sim C$  positions, which is at the energetically unstable ones. Their presence results in the appearance of two structural clusters (of fractional SOF value) at both sides of layer 17 for the  $\beta$ -phase.

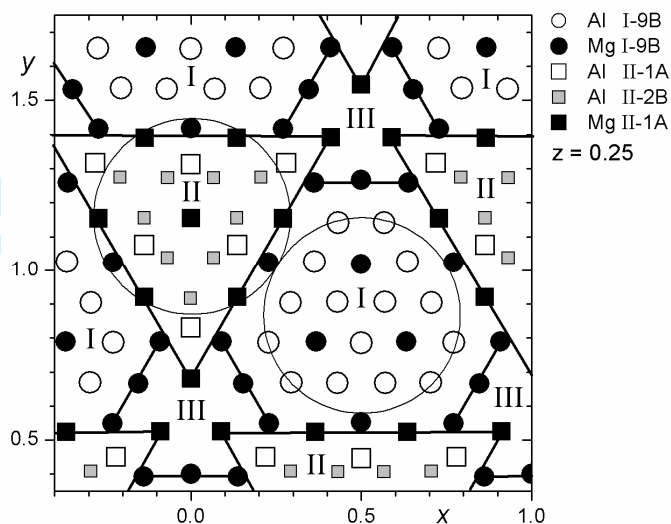


Figure 6. A cross-section of  $Mg_2Al_3$  structure for  $z=0.25$ . All atoms presented in the figure occupy lattice's knots of the domains. Domains I (layer 9B) and II (layers 1A and 1B) grow at the expense of domain III. In the presented cross-section, domain III vanishes – it loses its atoms. Light symbols denote Al atoms and black ones – Mg atoms.

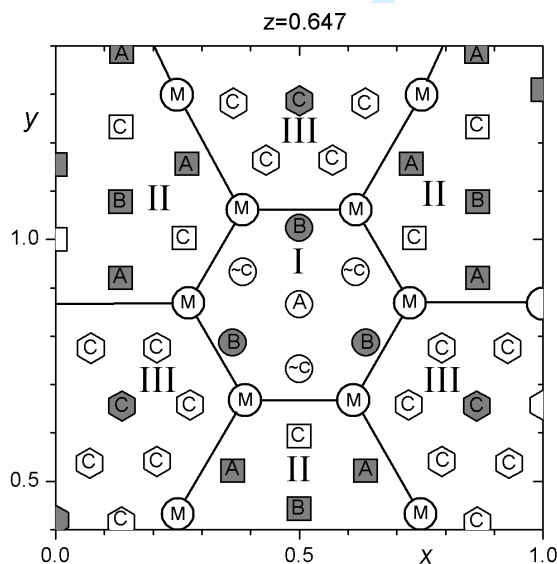


Figure 7. A cross-section of  $Mg_2Al_3$  structure for  $z=0.647$  (I-17' position). All the domains are occupied by Al (open symbols) and Mg atoms (full symbols). Domain I contains atoms positioned at  $\sim C$  knots, i.e. points at which clusters form when the temperature rises and the  $\beta$ -phase appears.

The last cross-section analyzed ( $z=0.383$ ) is presented in Figure 8. Only two domains (I and III) are marked in this figure; domain II is not shown. The cross-

section shows the transitional area of domain I, i.e. between the main chain of eleven layers and one of the shorter chains described by three layers 13'-15'. The figure describes a situation in which domains I and III grow into each other. Al atoms taking M-positions in domain I penetrate the inner area of domain III. Likewise, Al atoms positioned at M-positions of domain III can be found within the inner area of domain I. A similar cross-domain atomic penetration can also be observed at  $z=0.75$  for domains I and II.

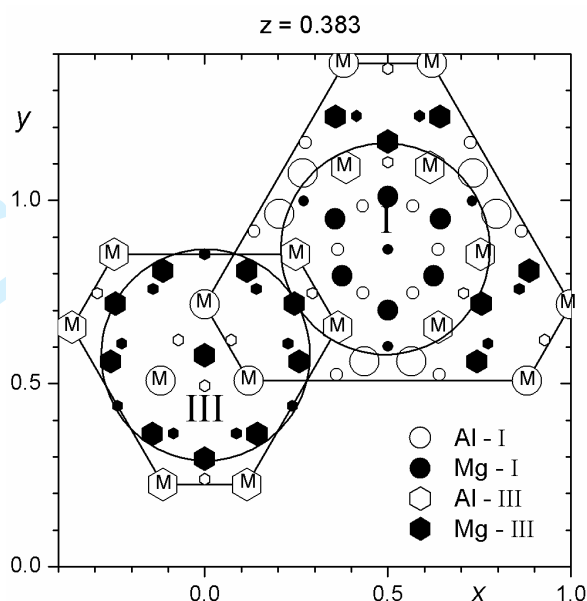


Figure 8. At  $z=0.383$  domains start to overlap each other. The circles ( $R = 0.29$ ) indicate the borders of the areas belonging to the particular domains. Al-atoms occupying M positions penetrate the neighbouring structural domains.

#### 4. Conclusions

In this paper the stable configurations of atoms for the closed-packed structures was discussed. The FCC or HCP local configurations are mostly observed for the packing of atoms with a hard-spheres interaction. In the case of the Lennard-Jones interaction, the BCC local arrangement of atoms also becomes stable. All those three configurations play an important role in the structural analysis of  $Mg_2Al_3$  compound for its high- and low-temperature phases: cubic- $\beta$  and rhombohedral- $\beta'$ . In order to compare these two structures, the unit cells coordinates were expressed in a hexagonal lattice. There is a hexagonal superstructure in the base plane, the periodicity of which is  $22/3 \approx 7.3$  times larger than the hexagonal lattice constant for Al atoms in a single domain.

The  $Mg_2Al_3$  structure consists of three types of rod-like hexagonal domains arranged in the sequence typical for a FCC-structure. The core of each domain contains the main chain of 11 hexagonal layers. The building element of the structure consists of hexagonal rings of aluminium atoms completed with dimmers of magnesium atoms as shown in Figure 3. The connection between complexes is carried out by hexagonal Al layers for which every second atomic position is empty (all even layers in Figure 1). The resulting structure consists of a sequence of Al hexagonal layers (with holes) separated by the appropriate Mg layers. An arrangement of such

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2  
3 building blocks can be also described by Friauf polyhedra, which are very common  
4 for the Laves phases: C14 and C36. The set of 11 layers does not undergo any  
5 transformation between  $\beta'$  to  $\beta$  phases. The most important problem is, however, the  
6 concentration of the core of the eleven layers. The corresponding value of the  
7 concentration is  $MgAl_2$  rather than  $Mg_2Al_3$  and the magnesium atoms are in  
8 deficiency. The Mg atoms are mostly located in the outer part of the domains.  
9  
10

11 The neighbouring structural domains should merge in a coherent manner.  
12 Three different mechanism of such a merging have been discussed for  $Mg_2Al_3$ . The  
13 first one occurs when the atoms occupy only A, B, C or M positions within two  
14 domains. The appearance of the third domain causes structural defects. This  
15 mechanism is shown in Figure 7 for layer 17' where some Al atoms occupy  $\sim C$  type  
16 positions which lead to instability and, in consequence, an atomic cluster formation  
17 observed in the higher-temperature phase.  
18  
19

20 The second example of a domain's arrangement is shown in Figure 6, where  
21 two domains (domain I – layer 9B and domain II – layers 1A and 2B) enlarge at the  
22 expense of the third domain, the size of which is reduced. Similarly to the case  
23 described in the previous paragraph, we observe a coherent merge between domains I  
24 and II whereas domain III is empty.  
25

26 The third way of merging is the mutual atomic intergrowth of two domains.  
27 This is outlined in Figure 8 for domains I and III. This process leads to a structural  
28 disorder observed in the higher-temperature phase and is manifested by an atomic  
29 shift with respect to their initial positions.  
30

31 The Mg atom occupies the centre of the alumina rings for  $\beta$  phase clusters.  
32 The resulting local structure is reminiscent of the BCC rather than the FCC structure.  
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34

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38 University of Science and Technology.  
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