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Precipitate/Matrix Interface in an Al-Mg-Ge Alloy Studied by Annular Dark-Field Scanning Transmission Electron Microscopy

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Precipitates in an Al-0.59Mg-0.71Ge (at.%) alloy similar to the trigonal U1-$MgAl_2Si_2$ phase in the ternary Al-Mg-Si system have been studied using aberration-corrected high-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM). The U1 phase forms as coarse needles with the needle direction along $<001>_{Al}$ in overaged Al-Mg-Si alloys. The structurally similar Ge-containing U1-like precipitates in the present alloy are typically smaller, have a different orientation in the cross-section plane and are present in considerable numbers close to peak hardness. The small U1-like precipitates are also more coherent with the Al matrix than in Al-Mg-Si alloys. They are generally lath-shaped, with cross sections elongated along one $<100>_{Al}$ direction. The investigation reveals the presence of a complex interface structure of Ge columns that appear at positions where the precipitate/matrix interface would otherwise have too short inter-atomic distances.

Keywords: *Al-Mg-Ge alloy, Al-Mg-Si alloy, STEM, aberration correction, precipitation.*

1. Introduction

Al-Mg-Ge alloys have been shown to share similarities with the industrially important Al-Mg-Si(-Cu) alloys [1-3]. Some of the precipitate phases known from the Al-Mg-Si system form isostructural phases in Al-Mg-Ge, specifically the trigonal U1 phase (a=b=4.05 Å, c=6.74 Å, γ=120°) [4] (also known as type-A or β'_A [5]) and the hexagonal β' phase (a=b=7.15 Å, c=4.05 Å, γ=120°) [6]. These two phases play a more important role in precipitation-hardening in the Al-Mg-Ge alloys than in the Al-Mg-Si system [1].

The U1-like phase has recently been investigated using non-aberration-corrected high-resolution transmission electron microscopy (HRTEM) [1]. This precipitate is needle-shaped, growing in a $<001>_{Al}$ direction and coherent with the matrix in this direction. However, the U1 precipitates are typically finer and produce higher hardness in the Al-Mg-Ge alloy than in Al-Mg-Si. The needle cross section is typically elongated along one $<100>_{Al}$ direction, with an almost planar interface between precipitate and matrix in this elongation direction. The c-axis of the U1 unit cell in most of the precipitates was found to lie close to $<210>_{Al}$. This is different from U1 in Al-Mg-Si, where the c-axis is parallel to $<310>_{Al}$ and the cross section is not elongated.

Several questions remained after this study. The orientation of the atomic contents of the U1 unit cell with respect to the precipitate/matrix interface could not be determined due to limited resolution, i.e., it was not possible to distinguish between two mirror images of the structure. Also, HAADF STEM images showed an increased intensity at the interface protruding into the matrix at semi-regular intervals. A better knowledge of the precipitate/matrix interface could explain why the

U1 phase behaves differently in the Al-Mg-Ge alloy. The precipitate/matrix interface appears to play an important role in determining the properties of the precipitate. Knowledge of the bulk structure of the precipitate is therefore not sufficient.

The strong atomic number dependence of HAADF STEM images makes it possible to distinguish between germanium (Z = 32) columns and magnesium or aluminium (Z = 12, 13) columns. Furthermore, intensity peaks in the HAADF STEM image correspond to atomic column positions, which makes the interpretation of these images less ambiguous than for example HRTEM images [7]. This is particularly important when looking at interfaces and defects [8].

2. Methods

The Al-0.59Mg-0.71Ge (at.%) alloy was cast and homogenized for 4 hours at 550 °C, before being extruded. The alloy was subsequently solution heat treated for 2 hours at 600 °C, water quenched to room temperature and held for 4 hours, followed by aging at 200 °C for 16 hours and water quenching to room temperature. TEM samples were prepared by twin-jet electropolishing with a nitric acid/methanol solution at -20 °C with a voltage of 13 V. HAADF STEM images were acquired on an FEI Titan^3 80-300 TEM at Monash University operating at 300 kV and equipped with aberration correctors (CEOS GmbH) for both the probe-forming and image-forming lenses. The probe convergence semi-angle in STEM mode was 15 mrad. The inner angle of the HAADF detector was 40 mrad and the outer angle on the order of 200 mrad.

3. Results and Discussion

An unprocessed HAADF STEM image of a U1-like precipitate viewed along $<001>_{Al}$ is shown in Fig 1(a). A low-pass filtered version is shown in Fig. 1(b). Spatial frequencies beyond the outermost Fourier-space peak were excluded in order to reduce the amount of noise. An enlarged view is given in Fig. 1(c). The Ge columns appear as the brightest spots in the precipitate. Rows of Mg columns are also clearly resolved in most of the precipitate. These form a light band in between the rows of Ge columns. The Al columns of the precipitate can be seen as intensity extending out from the Ge columns. The space between the Al columns forms a darker band. This atomic configuration, superimposed in Fig. 1(c), is in agreement with the model [1, 4]. The orientation of the atomic contents of the unit cell with respect to the matrix has thus been determined. The mirror image of the superimposed unit cell (i.e., the unit cell flipped about the c-axis) does not fit this precipitate. Most U1-like precipitates observed in the analysed condition had the same orientation.

In the enlarged view of a segment of the precipitate/matrix interface in Fig. 1(d), it can be seen that the Al columns are nearly aligned with the cross-section elongation direction. The $(011)_{U1}$ planes are parallel to the $(200)_{Al}$ planes of the matrix. This orientation makes the Ge columns also aligned with the $<100>_{Al}$ direction of the matrix. The precipitate is terminated on the left and right side by a zigzag line of Ge columns.

There is no large-scale deformation of the matrix surrounding the precipitate. A short-range displacement of the matrix columns is however visible right at the interface. The interface at the pointy ends of the precipitate is more diffuse. This interface probably moves more quickly during growth, and there is less coherence. Some of the matrix columns at these ends are brighter than the matrix columns in the bulk. This could be due to the presence of Ge atoms on fcc matrix positions.

A noticeable feature of the image is the bright columns appearing at the interface at semi-regular intervals. These extra columns most likely consist of germanium. They do not lie on matrix positions nor are they a continuation of the precipitate structure. They are specific to the precipitate/matrix interface. They are most common where the zigzag line of Ge columns points into the precipitate, although they can also appear (slightly dimmer) where the zigzag line points toward the matrix. The periodic nature of the appearance of these extra Ge columns suggests that they are not randomly

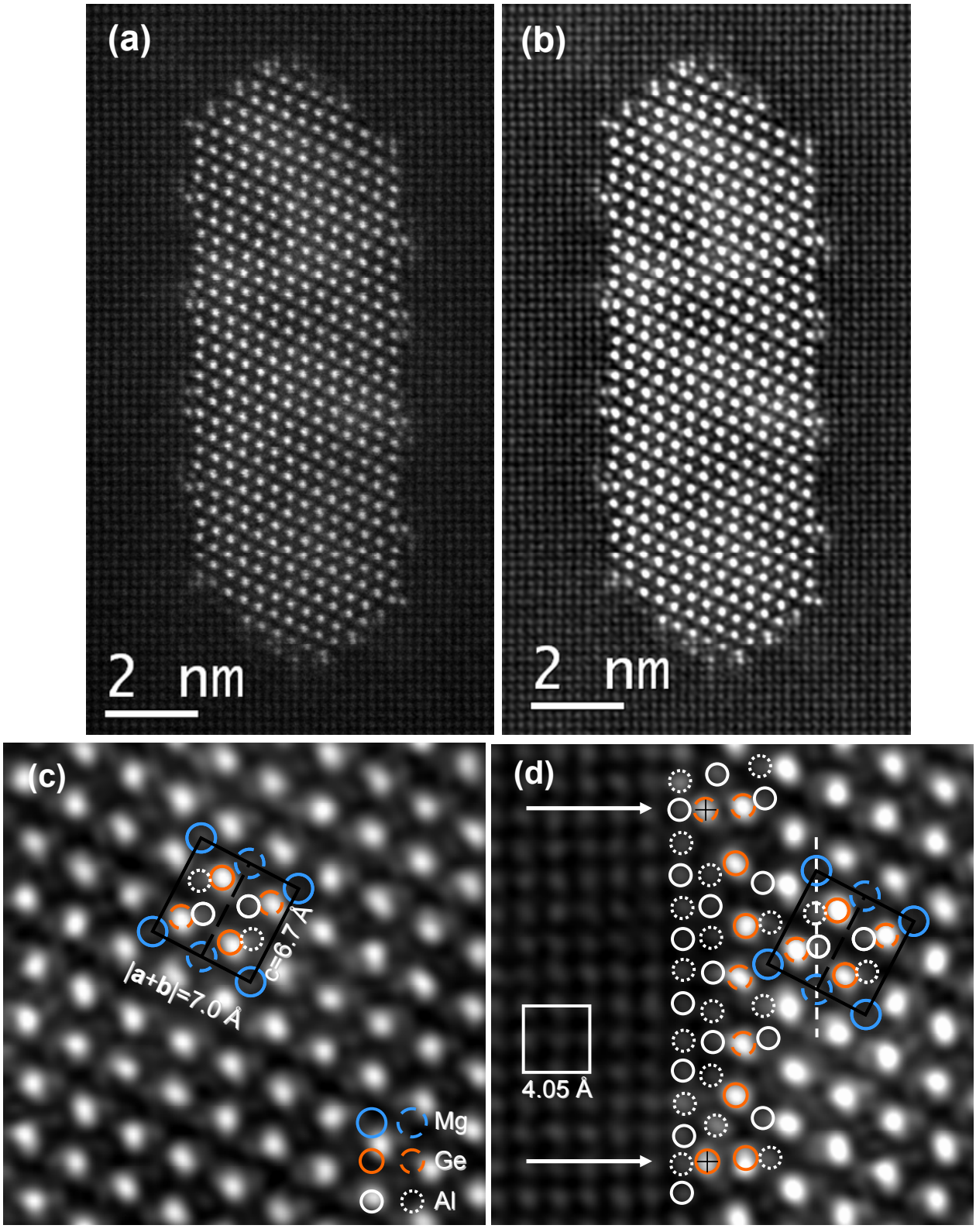

Fig. 1: (a) Unprocessed HAADF STEM image of a U1-like precipitate in the AlMgGe alloy. (b) Same image after low-pass Fourier-filtering. The brightest pixels have been clipped in order to make the weaker atomic columns more visible. (c) Enlarged view of the U1-like precipitate with a U1 atomic model superimposed. The full and dashed circles represent atoms at different heights. (d) Enlarged view of the precipitate/matrix interface. The zigzag line of Ge columns at the interface and the fact that the Al columns of the precipitate form a nearly straight line parallel to the interface can be seen. The vertical dashed line is a trace of the $(011)_{U1}$ plane. Arrows point towards the extra Ge columns on the interface (crossed).

distributed. We propose a possible explanation for the extra Ge columns by considering the staggering of the atomic columns.

Both the U1 precipitate and the Al matrix can be described as consisting of columns along the needle (i.e., viewing) direction made up of atoms separated by 4.05 Å, the lattice parameter of Al and U1 in this direction. The columns are staggered so that neighbouring columns are offset by 2.025 Å along the needle direction. Hence, next-nearest neighbours have atoms at the same height (for convenience, this will be referred to as the columns being at the same height). At the precipitate/matrix interface, one would expect little structural deformation when a precipitate, e.g., germanium, column and the neighbouring matrix column are at different heights. Based on this assumption, we can superimpose an atomic model on the columns at the interface in Fig. 1(d). When neighbouring columns are at the same height, one would expect something to give. We observe in the figure that the extra Ge columns only occur where the matrix column would have been right next to a precipitate Ge column at the same height. The extra Ge columns appear most often where the zigzag line points into the precipitate with a periodicity of 8 or 11 $(200)_{Al}$ planes (counting along the interface between two columns where the zigzag line points into the precipitate). These extra columns also appear where the zigzag line points toward the matrix. Although in this case, the extra column of Ge atoms is less bright, or there is neither a Ge column nor a matrix column visible, as in Fig. 1(d).

A missing matrix column can be explained by a lack of space. The column could also be partially occupied or the column could be strained (i.e., the lateral position of the atoms varies down the column), which can lead to a reduced HAADF signal [9]. That a matrix column should be replaced by a Ge column is more surprising. It would seem that this would not remove the problem of neighbouring columns at the same height. However, the extra Ge columns are not on matrix positions, but instead farther from the precipitate (in projection) than the matrix columns at other parts of the precipitate/matrix interface. There might therefore be enough room to place an extra Ge column at the interface right next to a precipitate Ge column at the same height. The extra Ge columns often have less intensity than the Ge columns in the bulk of the specimen. This could be due to partial occupancy, strain, dechanneling onto lighter neighbouring columns or a combination these. The fact that the extra Ge columns appear where neighbouring precipitate and matrix columns otherwise would have been at the same height suggests that the superimposed atomic model is correct, and that this effect is due to the lattice mismatch.

The same interface structure was observed for all U1-like precipitates with the same orientation relationship in the analysed condition. Figure 2 shows a HAADF STEM image of a somewhat coarser U1-like precipitate. Here, the extra Ge columns more often come in pairs, one where the zigzag line of Ge columns points into the precipitate and right next to it where the zigzag line points toward the matrix. We observe that the zigzag line at the interface is straighter than in the bulk of the precipitate. The precipitate structure is therefore deformed at the interface. The spacing between the extra Ge columns where the zigzag line points into the precipitate is, again, always 8 or 11 $(200)_{Al}$ planes.

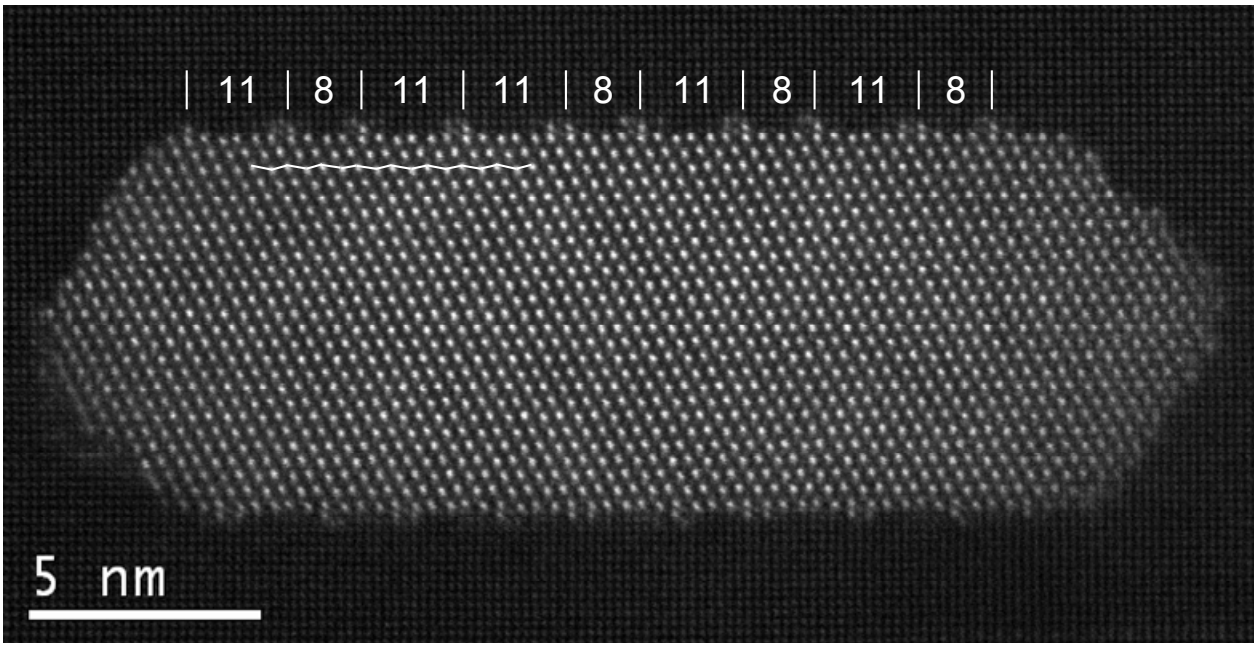

Fig. 2: Unprocessed HAADF STEM image of a U1-like precipitate from the same condition. The zigzag line formed by the Ge columns at the interface is not as uniform as that in the bulk of the precipitate (shown). The vertical lines indicate the extra Ge columns where the zigzag line points into the precipitate. The numbers indicate the number of $(200)_{Al}$ planes between the lines.

4. Conclusion

The U1-like precipitates in an Al-0.59Mg-0.71Ge (at.%) alloy were investigated using aberration-corrected HAADF STEM. Most of these needle-shaped precipitates had cross sections elongated along $<100>_{Al}$ with $(011)_{U1} \parallel (200)_{Al}$. This orientation aligns rows of both aluminium and germanium columns in the precipitate with the surrounding matrix, leading to a partially coherent interface. The precipitate is terminated at the interface by a zigzag line of Ge columns. Due to the structural mismatch, the interface cannot be perfectly even. Where the atoms of two neighbouring columns would have been at the same height and the zigzag line points toward the matrix, a matrix column is either missing or an extra column of germanium forms at the interface. Where the atoms of two neighbouring columns would have been at the same height and the zigzag line points into the precipitate, an extra column of germanium seems to always form at the interface.

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