Aberration-corrected HAADF-STEM investigations of precipitate structures in Al-Mg-Si alloys with low Cu additions

Takeshi Saito[1], Calin D. Marioara[2], Sigmund J. Andersen[2] Williams Lefebvre[3] and Randi Holmestad[1]

[1]Department of Physics, Norwegian University of Science and Technology (NTNU), N-7491 Trondheim, Norway
[2]SINTEF Materials and Chemistry, N-7465 Trondheim, Norway
[3]Université de Rouen, GPM, UMR CNRS 6634 BP 12, Avenue de I'Université, 76801 Saint Etienne du Rouvray, France

Abstract

Precipitates in a lean Al-Mg-Si alloy with low Cu addition (~0.10 wt%) were investigated by aberration corrected high angular annular dark field scanning transmission electron microscopy (HAADF-STEM). Most precipitates were found to be disordered on the generally ordered network of Si atomic columns which is common for the metastable precipitate structures. Fragments of known metastable precipitates in the Al-Mg-Si-(Cu) alloy system are found in the disordered precipitates. It was revealed that the disordered precipitates arise as a consequence of coexistence of the Si-network. Cu atomic columns are observed to either *in-between the Si-network* and *replacing a Si-network column*. In both cases, Cu is the center in a 3-fold rotational symmetry on the Si-network. Parts of unit cells of Q' phase were observed in the ends of a string-type precipitates known to extend along dislocation lines. It is suggested that the string-types forms by a growth as extension of the B'/Q' precipitates initially nucleated along dislocation lines. Alternating Mg and Si columns form a well-ordered interface structure in the disordered Q' precipitate. It is identical to the interface of the Q' parts in the string-type precipitate.

1. Introduction

Al-Mg-Si alloys are structural materials characterized by a high number density of precipitates yielding interfacial strain into the Al matrix and hindering dislocation movement. The efficiency of such obstruction depends highly on precipitate structure (type) because of different atomic bonding on the interface. The precipitates form with relatively low amounts of solute elements (up to 2 wt% of Mg and Si), which become supersaturated in the fcc Al lattice after solution heat treatment and rapid cooling. During a subsequent artificial aging, atomic clusters form and grow into metastable precipitates. The precipitation sequence of the Al-Mg-Si alloys is as follows [1-7]:

SSSS → atomic clusters → GP zones (pre-β") → β"→ β', U1, U2, B' → β, Si

where SSSS stands for super-saturated solid solution. In recent years the structures of the various metastable precipitates have been investigated in great detail by transmission electron microscopy (TEM) based studies; the main hardening precipitates are the highly-coherent, needle-shaped GP zones and β" which form the finest microstructure at peak hardness conditions

[2]. The U1, U2 and B' structures come in the shapes of larger, thicker needles/rods/laths and are also known as Type-A, Type-B and Type-C, respectively [8]. They mostly form together with β' (rod) upon over-aging [5]. They are semi-coherent and produce coarse microstructures with low strength. The equilibrium β (Mg_2Si) phase can be plate or cube-shaped. It often coexists with pure Si particles with diamond structure, for alloys with sufficiently high Si content.

The addition of Cu (~0.40 wt%) alters the precipitation sequence as follows [9-13]:

SSSS → atomic clusters → GP zones (pre-β") →β", L, S, C, QP, QC → β', Q' → Q.

This shows that Cu suppresses the formation of β", and causes other metastable precipitates such as L, S, C, QP and QC form at peak hardness conditions [9, 11]. Q' forms in over-aged conditions [9, 12]. This phase is most likely isostructural to the equilibrium Q phase [14] and to the B' phase in the Cu-free system [15].

In the Al-Mg-Si-(Cu) alloy system, all metastable precipitates, as well as the equilibrium Q phase, are structurally connected through a common network of Si atomic columns (the Si-network) with a projected near hexagonal symmetry of a = b ≈ 0.4 nm, $c = n \times 0.405$ nm (n is an integer), with *c* parallel to the needle/rod/lath directions [7, 9]. These precipitates are basically a stack of elemental columns in a <100>Al direction adapting the Al periodicity and can be defined as different arrangements of Al, Mg (and Cu) atomic columns situated in-between the triangularly arranged (≈ 0.4 nm spaced) columns forming the Si-network. The Si-network is distorted in the case of the β" phase as a consequence of the high coherency in this phase with the Al matrix.

In a previous work [16], we demonstrated that additions of low amounts of Cu (~ 0.10 wt%) to the Al-Mg-Si alloy system do not alter the precipitation sequence, although precipitation kinetics and number density of the precipitates change. In addition to perfect β" precipitates, which are typically encountered at the peak hardness conditions of Al-Mg-Si alloys, large fraction of precipitates consisting of perfect β" and disordered regions in the same precipitate needle (β"/disordered precipitates) was observed in the Cu-containing alloy. Using probe uncorrected HAADF-STEM, no Cu could be detected in the perfect β" parts while Cu atomic columns were observed within the disordered parts. Furthermore, it was found that the disordered parts are based on above described Si-network. It was suggested that Cu atomic columns are associated with two specific local symmetries on the network (atomic configurations) by Mg, Si and Al atomic columns connected to the Si-network: Cu column either *in-between the Si-network's columns* (being surrounded by Mg and Si atomic columns of the Si-network), or *replaces Si-network column*. While the exact atomic arrangement in the latter case could not be determined, it was suggested that the former local symmetry is similar to the one proposed earlier by Torsæter et al. [17]. This symmetry is not present in the bulk β" precipitate, which might related to its Cu-free composition [16].

Previous studies have reported that 10% pre-formation causes a heterogeneous precipitate distribution, with formation of different precipitate types in Al-Mg-Si alloy system: string-like (the 'string-type') and roundish (B'/Q' precipitates) cross-sections along dislocation lines [16, 18-21]. A low Cu addition (~ 0.10 wt%) preserved these types, although Cu atomic columns were observed in all investigated precipitates, as confirmed by probe uncorrected HAADF-

STEM [16]. It was suggested that these precipitates contain the same specific local symmetries surrounding Cu atomic columns. However, the atomic structures in details could not be determined due to the limited resolution of the HAADF-STEM instrument [16]. Hence, discussion of the atomic column symmetries and its connection to the Si-network were yet hypothetical. In this work, we study the local atomic structure of these precipitates in details. Aberration-corrected HAADF-STEM, used for the results presented in this work, enables to determine Cu local symmetries, structure of the disordered parts of the precipitates and their interface structure with the Al matrix. The new knowledge can contribute to improve understanding of precipitation and growth mechanism and potentially to aid microstructure modeling in its efforts to connect micro-/nano-structure to macroscopic properties of the alloys.

2. Experimental procedure

An alloy with chemical composition of 0.47 Mg, 0.42 Si, 0.07 Fe and 0.10 Cu (wt%) was used for all experiments. The composition was measured by inductively coupled plasma optical emission spectroscopy. Other impurities had a total less than 0.01 wt%. The alloy was cast as a cylindrical ingot, from which the extrusion billet was cut. The billet was homogenized at 575°C for 2.5 hours and subsequently extruded to a round profile with 20 mm in diameter. The extrusion was conducted well above the solvus temperature for the alloy. From the extruded profiles, the alloys were machined with tensile test geometry for making a predeformed condition while others were cut transversally for an undeformed condition. All alloys were solution heat treated (SHT) in a salt bath at 545°C for 5 minutes, and subsequently water quenched. They were then stored at room temperature (i.e. natural aging) for a total of 30 minutes. The predeformed conditions were made by plastically deformed to 10% by tensile stress within 5 minutes after the SHT (during the natural aging). All alloys were further heat treated isothermally (i.e. artificial aging) at 190°C for 300 minutes. The preparation and thermo-mechanical history are identical to the previous work, which are fully described in [16].

HAADF-STEM specimens were prepared by electropolishing using a Tenupol 5 machine (Struers), on transversal slices along the extrusion and deformation directions in the case of undeformed and predeformed conditions, respectively. The electrolyte consisted of 1/3 vol% HNO_3 in methanol and the solution was kept at a temperature between -20°C and -35°C.

The HAADF-STEM images were taken by a spherical aberration probe corrected JEOL JEM-ARM200F TEM with a Schottky field emitter operated at 200 kV. The probe diameter was 0.1 nm and the collection angle of the HAADF detector was in the range of 45-150 mrad. The HAADF-STEM technique enables to determine atomic column positions directly, being less affected by objective lens defocus and specimen thickness compared to high resolution TEM images. In addition, the technique provides atomic number (Z) contrast with the intensity proportional to $Z^{1.7\text{-}1.9}$ [22, 23]. This enables to distinguish the heavier Cu (Z_{Cu} = 29) atomic columns from those of Mg (Z_{Mg} = 12), Al (Z_{Al} = 13) and Si (Z_{Si} = 14) in the precipitate structures. In order to reduce contamination on the specimen during the HAADF-STEM observation, all specimens were ion-milled by precision ion polishing system (PIPS Gatan) and plasma cleaned by plasma cleaner (SOLARUS Gatan) before the observation.

3. Results and discussion

All HAADF-STEM images shown in this paper were taken from material isothermally heat treated at 190°C for 300 minutes, which is known to produce a peak aged condition in this alloy [16]. Since the precipitate needles are growing in a <001>Al directions, all images were taken from the <001>Al zone axis, corresponding to the cross sections of the precipitate needle. Owing to the high resolution of aberration-corrected HAADF-STEM, all atomic columns could be resolved in the structures. Based on their high Z contrast, Cu atomic columns could be identified even in the unprocessed HAADF-STEM images. Fast Fourier transform (FFT) filtering has been applied to reduce noise using a circular band pass mask that removed all period shorter than 0.15 nm. After filtering the images, weaker-contrast Si, Al and Mg atomic columns could also be resolved. The atomic overlays for all figures in this paper were based on the Z contrast, inter-atomic distances and local similarities with well-known structures in the Al-Mg-Si alloy system. The following sub-sections discuss each type of precipitates observed.

3.1 β”/disordered precipitate

Figure 1 shows a typical example of the β”/disordered precipitate. It is here straight forward to distinguish between the perfect β” and the disordered parts in the precipitate. In this type of precipitates, the Cu atomic columns are found only within the disordered parts or at the precipitates/matrix interface, but never inside the β” structure. This observation supports results of the precious work [16]. It is interesting to note that the disordered part of the precipitate consists of several identical local atomic columns arrangements, and it appears to have no overall unit cell periodicity. These arrangements can be identified to be sub-cell fragments in different types of the other metastable precipitates in the Al-Mg-Si(-Cu) alloy system, such as C, Q', β'_{Ag} and U2 see Figure 1 and Figure 2. Figure 2 shows schematic drawing of unit cells of the encountered metastable precipitates. The legend in Figure 2 represents all figures in the present study. In the investigated precipitates, the disordered part is always based on the same ordered Si-network, see Figure 1. In addition, Cu atomic columns could be classified by their two different positions connected to Si-network: *in-between the Si-network columns* and *replacing Si-network columns*. The first case is identical to C, Q', Q and L precipitates [17]. This is dominant in respect to the fraction of Cu columns found in such positions. The second case is interestingly analogue to Ag columns in β'_{Ag} precipitates [24], but with Ag being replaced by Cu, see Figure 1 and 2. This local atomic configuration will be referred to as modified β' in the present study, and represents a structure which is isostructural with the β'_{Ag}. It is worth noting that both cases have 3-fold rotational symmetry around the Cu atomic columns in this projection, namely along needle growth direction. It seems plausible that Cu occupies only the sites that can produce 3-fold symmetry, or it modifies the surrounding columns to produce this type of symmetry. The β” structure lacks this type of sites (atomic columns) because it is more closely adapted to the matrix, with a maximum 2-fold symmetry, as given by its space group C2/m (12) [2, 3]. Cu is therefore only able to exist at the disordered part and interface. It is worth mentioning that Al and Mg atomic columns also take positions in-between the Si-network columns, however there is never a 3-fold rotational symmetry around them.

In a previous work [16], it was measured that Cu-added alloy (~0.10 wt% Cu) produces twice the precipitate number density than the base alloy (~0.01 wt% Cu). It seems therefore that Cu atoms exert a strong effect on nucleation; either they create additional nuclei, or they merge with the ones already present, improving potently nucleation. Based on above mentioned observations,

Cu atoms might produce clusters with atomic surrounding having 3-fold symmetry connected to the Si-network during nucleation. This hypothesis will be discussed in later sub-section.

As observed in Figure 1 and 2, some parts recognized to be associated with the U2 phase could frequently be observed in Cu-free areas of the disordered part of the β”/disordered precipitate. For instance, a long band consisting of the U2 structure acts as an interphase between bulk β” and the disordered part. It is interesting to mention that β” phase has fragment of U2 phase, see Figure 2 (a). This could be a reason why the band consisting of the U2 structure act as an interphase between bulk β” and disordered parts in β”/disordered precipitate. Similar U2 bands connect domains of β'_{Ag} in the precipitates of the respective system [24]. From Figure 1, it can be seen that the connection between the U2 band and the bulk β” is much accurate compositionally if the Mg1 site in β” phase is occupied by an Al atom. Therefore, a composition of Mg4Al3Si4 is chosen for the β” phase, which is one of the two lowest formation enthalpy structures according to previous study [3].

Cu atomic columns are also present to a certain extent at the precipitate/matrix interface, especially in the disordered part of the precipitate, see Figure 1. At the interface Cu atomic columns are spread unevenly. Since the interface is rather incoherent, there will be sites that are more and less well suited for the Cu atoms. Another plausible explanation is that these columns formed in defect sites created due to the interface growth during nucleation/precipitation. Further experiments are needed to understand the possible role of Cu taking in shaping the interface. There was only one Cu-enriched column observed at the interface of the bulk β” with the Al matrix, located on a Si3 site [3], see Figure 1. For the small area of the disordered part where the Si network is oriented along <010>Al (see the lower-right side of the precipitate in Figure 1), these three Cu atomic columns at the interface resemble local parts of C or L phase precipitates [17].

As mentioned, fragments of the C, Q’, β'_{Ag} and U2 phases were identified in the disordered parts. The Si-network is near-hexagonal in all these phases, but has two orientations (rotations in the (001) Al plane), which sets the orientation of the precipitate cross-section and interface directions. For the C and L phases, one vector of the hexagonal base of the Si-network is oriented along <100>Al. For the other phases, it is along <510>Al. This creates a discontinuity (faults) in the Si-network at the interconnected region of these components, as seen in Figure 1.

3.2 String-type and disordered Q’ precipitates along dislocation lines

Examples of the string-type and disordered Q’ precipitates are shown in Figures 3 and 4. It is revealed that both precipitate types are also composed of fragments from a number of different metastable precipitates in the Al-Mg-Si(-Cu) alloy system. Both types of Cu local configurations, with Cu column *in-between the Si-network* and replacing *on the Si-network column*, were observed in the string-type precipitate, as it was the case in the disordered part of the β”/disordered precipitates. Fragments of U2 phase are also easily identified. From these observations, we can conclude that all disordered precipitates are composed of parts of various metastable precipitates structure on a common near-hexagonal Si-network in some manner. It is interesting to note that half unit cells of Q’ precipitates were observed at the narrow ends of the cross-sections in the string-type precipitates, see Figure 3. It is well known that Q’ and the string-type precipitates preferably form along dislocation lines [16, 18-21]. This could indicate that Q’ precipitates (or fragments thereof) initially form along the dislocation line, and only later become connected by the string-type. For instance, for the precipitates in Figure 3, the half unit cell may

represent the Q' phase parts first nucleated. In other words, the string-type precipitates could start at different positions from small regions of Q' building out the Si-network which should encounter somewhere in-between. This also means that they start not only at arbitrary heights, but also they meet in wrong directions. This will interrupt the ordering the Si-network columns height, but could also lead to faults in the projected Si-network, both which will be sources of disorder. Note that the Si-network contains discontinuities in the non Q' precipitates regions, see Figure 3. It is interesting to note that another fragment was observed as shown by thick solid red line in Figure 4 (iii). The fragment is also connected with the ordering Si-network and is repeated like unit cell along <510>Al direction. However, this fragment has not reported yet as an pure precipitate phase. This could give interesting view to explain that Si-network might be much important than ordered precipitate structure during formation of precipitates, which will be discussed in next sub-section.

Higher number density of precipitates was observed in the condition with the low Cu addition (~0.10 wt%) compared to the base alloy (~0.01 wt% Cu) in the predeformed condition [16]. This again supports the hypothesis for Cu atoms promoting nucleation in these materials.

It is worth noting that the precipitate interface structure of the Q' part of the disordered precipitate in Figure 4 is well defined, which is shown by thick solid blue line on the upper side of the atomic overlay in Figure 4. Cu atoms are not incorporated in the interface structure, which instead consists of alternating Mg and Si atomic columns. This interface structure is found to be identical in the Q' parts at the ends of the string-type precipitate, see Figure 3. The alternating Mg and Si atomic columns are also present at the interface of the lower side of the precipitates, although they are much disordered manner.

3.3 Hypothesis of Cu local configurations in connection with Si-network during nucleation

The disordered precipitates observed may provide new insight into the nucleation stage. The fragments which not continue to grow into bulk structures point out that the Si-network at some stage is more energetically favorable than the ordered precipitate structures. Note that the fragments of the other precipitates observed in the disordered precipitates were based on atomic columns along the whole needle-like precipitates. This means that the fragments grow side-by-side throughout the length of the precipitates. It is not possible to know the details during nucleation and in the early growth stage without dedicated experiment. However, it is plausible to say that the fragments based on Si-network would form during nucleation, in the early growth stage, before growing as longer needles. During the early growth, several precipitate phases could be started off from the same 'local Si network'. Then, it allows more than one structure or fragment to be possible across the initial cross-section. It is understandable that the Cu aggregate could play an important role considering nucleation. Since Cu seeks to be surrounded by Si atoms, it would be influencing the local order on the Si-network at early stage and could make clusters more potent for the Cu-containing phases.

4. Conclusions

The precipitate structures formed in an alloy with composition 0.47 Mg, 0.42 Si, 0.07 Fe and a low addition of Cu (~0.10 wt%) were investigated by aberration corrected HAADF-STEM. The results obtained are summarized as follows:

- A high fraction of the precipitates were classified as disordered.

- Disordered precipitates are composed of local atomic column arrangements consistent with fragments of different types of metastable precipitates in the Al-Mg-Si-(Cu) alloy system, existing side-by-side on a common near-hexagonal Si-network.
- Each Cu atomic column on the network is observed to be the center in two types of columns arrangements. In the most frequent arrangement, it takes the place between three Si-network columns. In the less dominating arrangement, Cu replaces a Si-network column.
- In both arrangements the Cu corresponds to a 3-fold rotational symmetry axis, as viewed along the needle growth direction.
- Half unit cells of Q' phase was observed at the ends of a string-type precipitate. It is suggested that the string-type originates from initially nucleated Q' precipitates/fragments which extend and meet each other along the dislocation line.
- A well-ordered interface structure consisting of alternating Mg and Si atomic columns was observed for the Q' phase. This was identical to the interface of the Q' precipices/fragments which extend and encounter each other along the dislocation line.
- The results of disordered precipitates indicate that the fragments are established early in the growth, possibly on small domains where the Si-network is established on just a few (001)Al planes.
- Cu plays an important role for precipitate nucleation. It is suggested that Cu atoms influence the cluster during nucleation, priming them for Cu-containing phases, which explains the increased precipitate number densities in the Cu-containing alloys. It is likely such cluster could contain the specific Cu arrangements as discussed.

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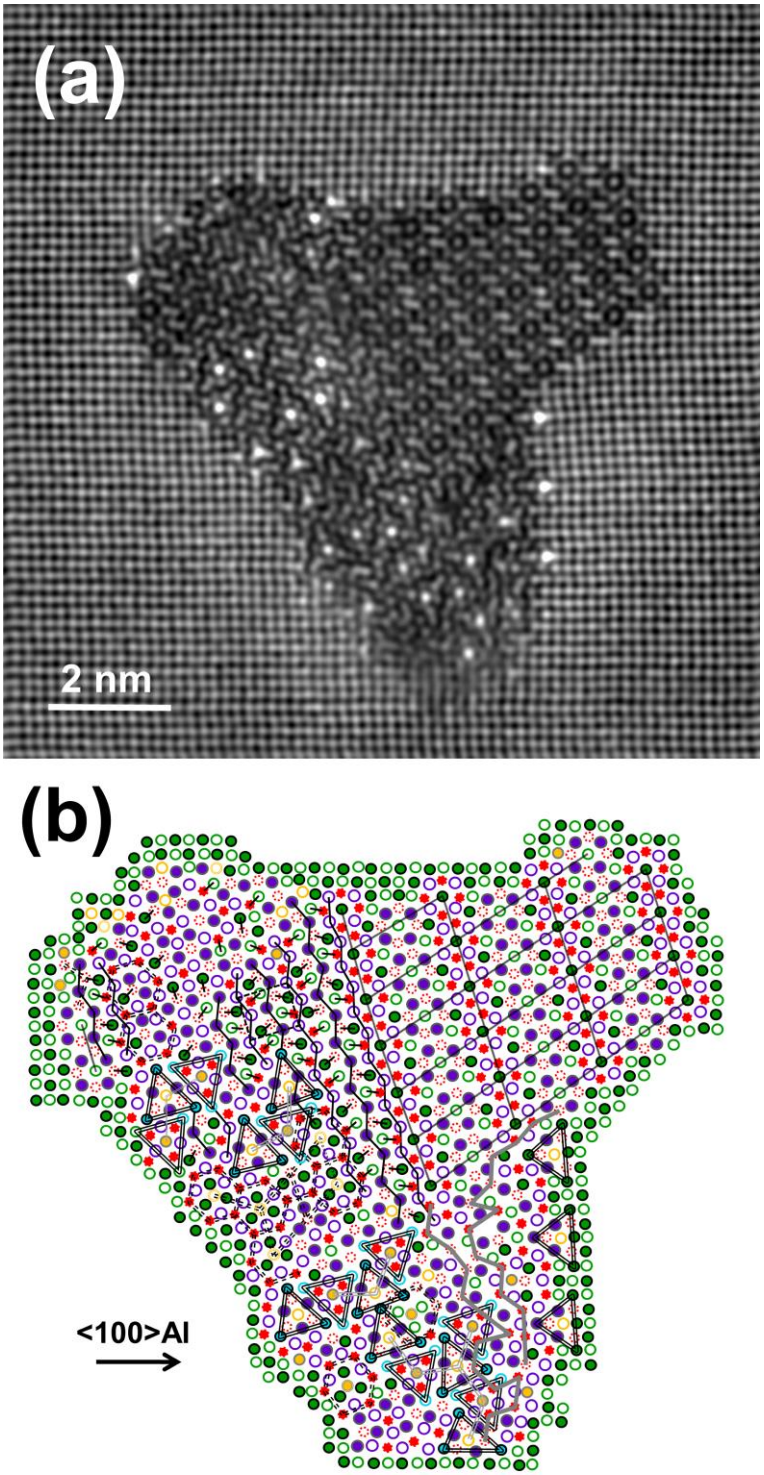

Figure 1 HAADF-STEM images of a β”/disordered precipitate cross-section taken along <001>Al for the undeformed condition. (a) Unprocessed image, (b) inverse FFT (IFFT) filtered image of (a). (c) Suggested atomic overlay. See legend in Figure 2.

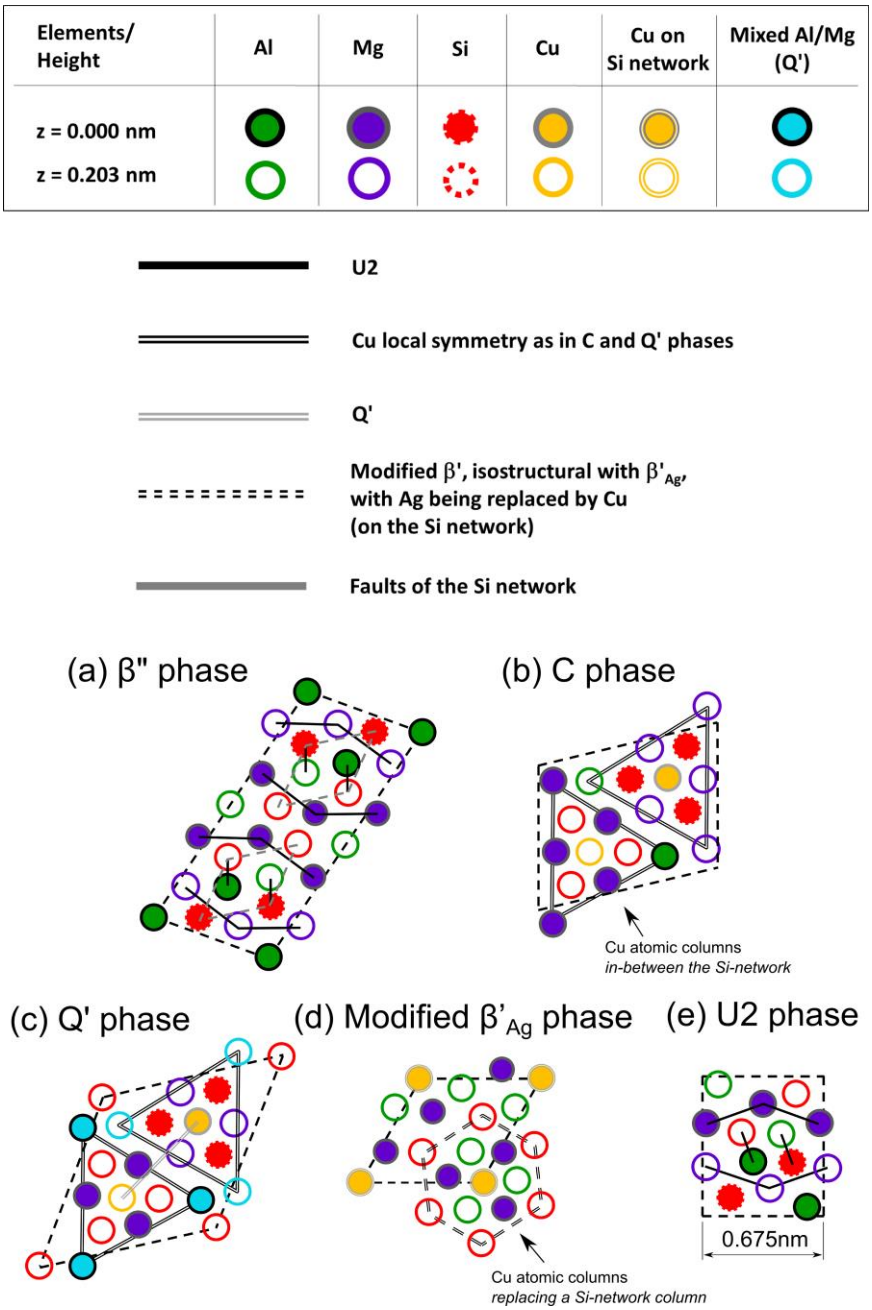

Figure 2 Legend representing all overlays in the present study and schematic drawings of unit cells of the encountered metastable precipitates: (a) β" phase [3], (b) C phase [17], (c) Q' phase [14], (d) modified β'_{Ag} phase [24] and (e) U2 phase [7], drawn to the same scale. The atomic column arrangement around Cu is shown for both *in-between the Si-network* and *replacing a Si-network column*. Cu atomic columns for the former case is identical to C, Q, Q' and L phases [17] and the latter case is found in β'_{Ag} phase [24] (Cu atomic columns instead of Ag in this case). Both Cu arrangements have 3-fold rotational symmetry. Neither exists in the β" phase probably because the space group of C2/m (12) at most allows 2-fold rotation although parts of the near-hexagonal Si-network is found [2, 3], see gray dashed line in (a). The β" phase has fragments of U2 phase, which can create interface between perfect β" and disordered parts along <310> Al direction in β"/disordered precipitate, see Figure 1.

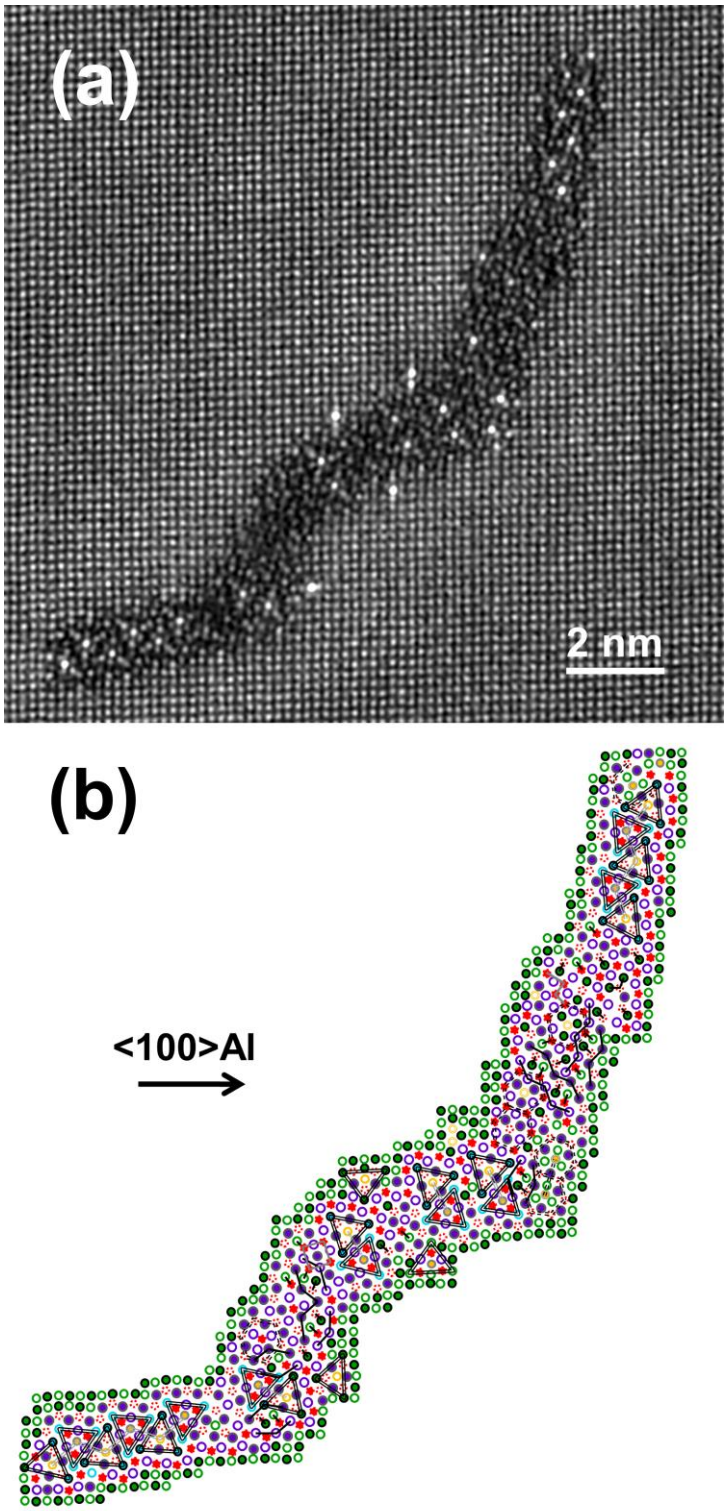

Figure 3 HAADF-STEM images of string-type precipitate cross-section taken along <001>Al for the 10% predeformed condition. (a) Unprocessed image, (b) IFFT filtered image of (a). (c) Suggested atomic overlay on the image (b). See legend in Figure 2. Half units cell of the Q' phase are present at both ends of the precipitate.

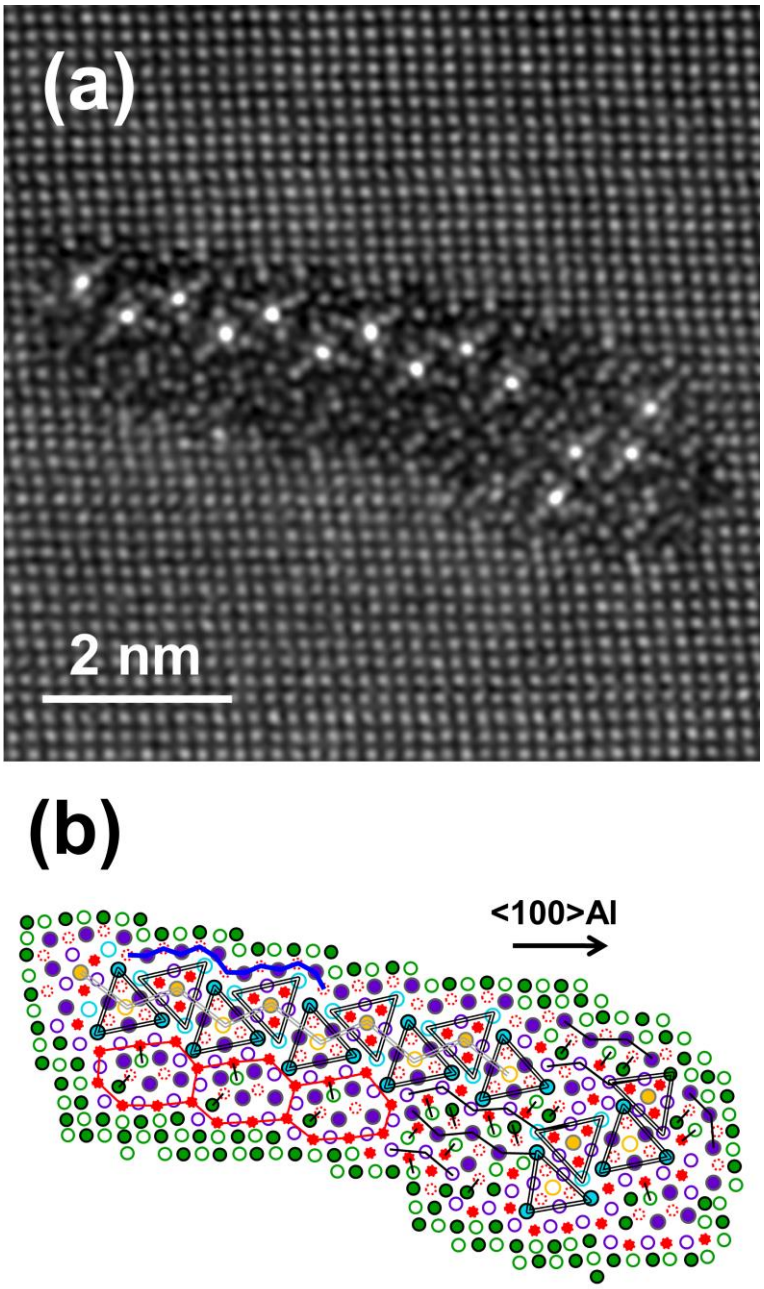

Figure 4 HAADF-STEM images of precipitate cross-section taken along <001>Al for the 10% predeformed condition. (a) Unprocessed image of disordered Q' precipitate, (b) IFFT filtered image of (a). (c) Suggested atomic overlay on the image (b). See legend in Figure 2. In addition, the alternating Mg and Si columns in the interface of the Q' part with Al matrix along <510> Al is shown by thick solid blue line. Another fragment, which have not been reported yet as a pure precipitate phase in the Al-Mg-Si system, are shown in thick solid red line.