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Imperfection of and phase transformation in Al–Cu–Mg quasicrystals

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Abstract. The rapidly solidified Al–Cu–Mg alloy is studied by electron diffraction at room temperature and under *in situ* heating. The distortion of electron diffraction patterns, which characterizes an intermediate state between crystalline and icosahedral quasicrystalline states, is interpreted by a particular quenched linear phason strain. It is shown that the phase transformation from the imperfect icosahedral quasicrystalline Al–Cu–Mg to crystalline R-$Mg_{32}(Al, Cu)_{49}$ is accompanied by a change in phason strain.

1. Introduction

Diffraction patterns of perfect icosahedral phase consist of sharp peaks and exhibit icosahedral point group symmetry $m\bar{3}\bar{5}$ [1]. However, some quasicrystal grains have been found to be imperfect quasicrystals characterized by splitting and shift of diffraction peaks. Generally, the weak peak has a large shift and the strong peak has a small shift [2]. It was pointed out that the splitting of diffraction peaks in fact results from the superposition of two or more diffraction patterns where the shift of the same peak occurs along different directions [3]; the non-delta-function electron diffraction spots and x-ray diffraction peaks have also been reported [4, 5]. Several research groups have suggested that the distortion of diffraction patterns is caused by quenched phason strain and the shift of diffraction peaks can be explained by a linear phason strain [2, 4–6]. It was shown by the precession x-ray diffraction result that the peak shift in diffraction patterns of T2-Al_6CuLi_3 leads to the degeneration of the symmetry from $m\bar{3}\bar{5}$ into $m3$ and this can be explained well by a particular quenched linear phason strain [7]. Li *et al* [8] reported that a series of electron diffraction patterns including slightly distorted, heavily distorted and crystalline-like diffraction patterns had been obtained for Al–Cu–Li alloy. These patterns are considered to correspond to different degrees of imperfection of quasicrystals. It was illustrated that the degree of imperfection of icosahedral quasicrystalline Al–Cu–Li alloy can be described by the degree of deviation of the quasilattice from its idealized form under the action of a linear phason strain, and the deviation is an approximation to the BCC lattice of R-Al_5CuLi_3 phase. In this sense, the crystalline phase R-Al_5CuLi_3 can be recognized as an extremely distorted icosahedral phase [8]. In addition, it was reported that the electron diffraction patterns of icosahedral phases would be distorted after annealing [9, 10]. This calls our attention to the connection between the phason strain and the phase transformation from quasicrystals to

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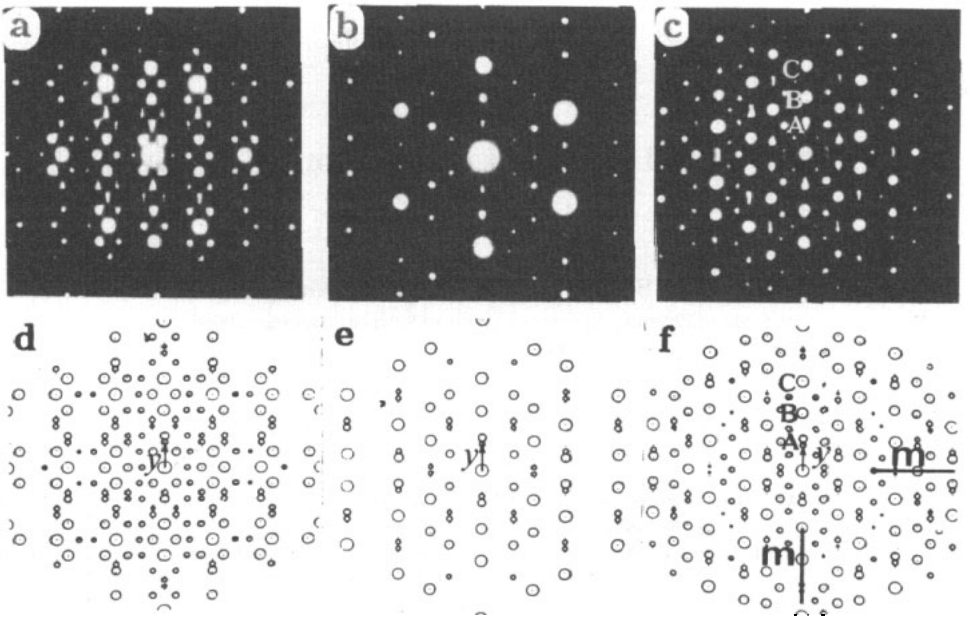

Figure 1. Electron diffraction patterns of rapidly solidified Al–Cu–Mg alloy taken along (*a*) twofold, (*b*) threefold and (*c*) fivefold axes. (*d*), (*e*) and (*f*) Calculated reciprocal-quasilattice planes corresponding to (*a*), (*b*) and (*c*), respectively, with the linear phason strain M_1 (b = 1.25).

crystals. In addition, a high-resolution electron microscopy study indicates that the rapidly solidified Al–Cu–Mg alloy may be in between the crystalline and quasicrystalline states [2]. Its diffraction patterns also deviate from the standard patterns of icosahedral quasicrystals but they are different from those of Al–Cu–Li alloy. This paper tries to show whether the intermediate state between crystal and quasicrystal observed in the Al–Cu–Mg alloy can also be described as an imperfect quasicrystal with a particular quenched linear phason strain, and whether the phase transformation from the imperfect icosahedral quasicrystalline Al–Cu–Mg to the crystalline R phase is related to the change in phason strain.

2. Experimental details

The homogeneous Al–Cu–Mg alloy was first formed in the vacuum from elements at 99.99% purity to give 47.6 wt% Al–19.9 wt% Cu–32.5 wt% Mg alloy. The melted alloy was rapidly solidified into thin ribbons by spinning with a single copper roller. The ribbon samples were thinned by ion milling and were cooled under liquid nitrogen during ion milling. They were observed at room temperature with a Philips EM-420 electron microscope and under *in situ* heating with a Philips EM-400 electron microscope.

3. Intermediate state between two-dimensional and three-dimensional quasicrystals

Figures 1(*a*), 1(*b*) and 1(*c*) are electron diffraction patterns of one quasicrystalline grain taken at room temperature along the degenerate twofold, threefold and fivefold axes,

respectively. In figure 1(*c*) the fivefold symmetry of the diffraction pattern is broken and degenerates approximately to 2 mm with the vertical and horizontal centre lines as lines of symmetry if some diffuse scattering is ignored. It can be seen that all spots almost align periodically along the vertical direction owing to the spot shift (especially the shift of the weak spots). Some adjacent spots displace towards each other and unite to form elongated spots, and the spot distance along vertical direction corresponds approximately to the spacing of the (200) plane in the crystalline $Mg_{32}(Al, Cu)_{49}$ phase (R phase), while the spacings among vertical reciprocal-lattice lines still obey the golden ratio. In figures 1(*a*) and 1(*b*), an approximate periodicity along the vertical direction and a similar elongation of diffraction spots to that shown in figure 1(*c*) can be found. The spacings between vertically aligned spot rows also obey the golden ratio. Hence, these diffraction patterns show the structural features of both a crystal and a quasicrystal. This phenomenon can be interpreted by a particular quenched linear phason strain $w(x) = xM$ [3]. M is a second-rank tensor and can be determined from the electron diffraction patterns shown in figure 1 because the shift amount of the diffraction peak equals the product of M and $G_\perp$; here $G_\perp$ denotes the component of the six-dimensional reciprocal-lattice vector in pseudospace [3]. M is chosen as follows:

$$M_1 = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -(\tau - b)/(\tau b + 1) \\ 0 & 0 & 0 \end{pmatrix}$$

τ represents the golden ratio. Here, we use the Cartesian coordinate system where the X, Y, Z axes coincide with three twofold axes of an icosahedron orthogonal to one another [11]. The elongation of diffraction spots in figure 1 is along the direction Y. The value of b can be determined from the ratio of distances between diffraction spots by the expression $b = \overline{CB}/\overline{BA}$, where A, B and C denote the diffraction spots as shown in figure 1(*c*). Obviously, when b equals τ, M_1 becomes zero and the phason strain vanishes. Figures 1(*d*), 1(*e*) and 1(*f*) are calculated reciprocal quasilattice planes with $b = 1.25$, which correspond to figures 1(*a*), 1(*b*) and 1(*c*) respectively. The small dots denote the reciprocal-quasilattice points for the perfect icosahedral quasicrystal. The open circles represent the shifted reciprocal-quasilattice points under the action of linear phason strain. The radii of the circles are proportional to the weight of reciprocal-quasilattice points. Evidently, figures 1(*d*), 1(*e*) and 1(*f*) correspond well to figures 1(*a*), 1(*b*) and 1(*c*), respectively. The misfit seen from the shape of some diffraction spots and the size of some weak spots may be due to intensity difference. Here the weights of reciprocal-lattice points are calculated for a perfect quasicrystal, and the decoration of atoms is not considered.

Figure 2 shows that, when $b = 1$, namely when the elements in M_1 equal $-(\tau - 1)/(\tau + 1) = -0.236$, all circles align along the vertical direction with a strict periodicity. It was illustrated that a similar phason strain, but spreading along three orthogonal directions, makes the icosahedral quasilattice transform into the BCC lattice of the R phase [7]. In this sense, figure 2 corresponds exactly and figure 1 corresponds approximately to a two-dimensional quasilattice or a one-dimensionally crystallized quasilattice. However, this kind of two-dimensional quasicrystal is different from the decagonal phase in Al–Mn and Al–Fe alloys [11, 12]. The decagonal phase has periodicity along a tenfold axis, while in figure 2 the periodicity is not along the direction of the fivefold axis but along one of the twofold axes, and in fact there is no fivefold rotational symmetry in this state. Generally speaking, one can always find a particular linear phason

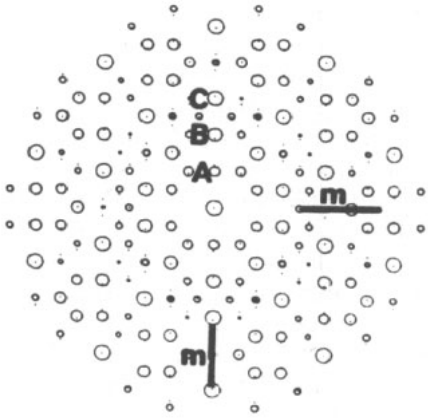

Figure 2. Calculated reciprocal-quasilattice plane normal to a fivefold axis with the phason strain $M_1(b = 1.0)$. The spots are arranged with a strict periodicity and quasiperiodicity along the vertical and the horizontal, respectively.

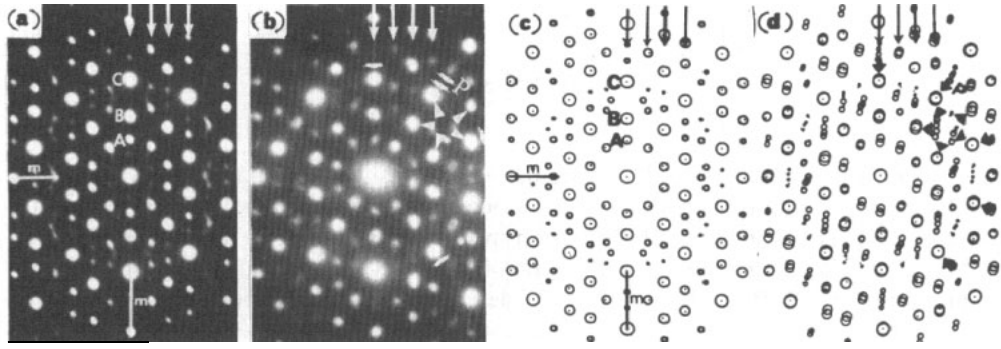

Figure 3. Electron diffraction patterns of rapidly solidified Al–Cu–Mg alloy taken during *in situ* heating (*a*) at room temperature and (*b*) at 284 °C (just before crystallization). (*c*), (*d*) Calculated reciprocal-quasilattice planes corresponding to (*a*) and (*b*), respectively (see text).

strain which makes the icosahedral quasilattice deviate from the idealized form and approach towards the ordinary crystalline lattice.

4. Crystallization and change in phason strain

In situ heating transmission electron microscopy observation has been carried out. The temperature was measured with a thermocouple placed close to the specimen on the heating stage. Up to a temperature of about 273 °C, no change is seen in both the diffraction patterns and the microscopic image of the specimen. At about 284 °C the icosahedral quasicrystalline grain transforms into a crystalline grain; the transformation starts from the edge of the specimen, and the whole icosahedral grain transforms quickly in the wake of the transformation front. Figures 3(*a*) and 3(*b*) are electron diffraction patterns taken at room temperature and just before the crystallization (284 °C), respectively. The distortion of reciprocal-quasilattice planes in figure 3(*a*) is that all spots are arranged in parallel and almost equispaced vertical lines as shown by arrows. Both the

horizontal and the vertical mirror planes still exist. In figure 3(*b*), besides the same distortion shown in figure 3(*a*), all spots align almost equidistantly along the direction labelled P, and the arrangements of spots along the five directions indicated by broad arrows are different from each other. This indicates that, after heating, the sample becomes a more perfect icosahedral quasicrystal, and the transformation process to a more imperfect quasicrystal is none other than the process to a more periodic state. These two diffraction patterns can also be simulated by introducing a linear phason strain with a particular form. Various distorted electron diffraction patterns have been investigated, and all of them indicate that the reciprocal-quasilattice points lying on x, y and z axes always shift parallel to the x, y and z axes, respectively. We assume that this is also true for this quasicrystal grain. Figures 3(*c*) and 3(*d*) are the simulated distorted reciprocal-quasilattice planes for figures 3(*a*) and 3(*b*), respectively. Figure 3(*c*) is calculated under the action of the phason strain expressed by

$$M_2 = \begin{pmatrix} (\tau - a)/(\tau a + 1) & 0 & 0 \\ 0 & 0 & -(\tau - 1.2)/(1.2\tau + 1) \\ 0 & 0 & 0 \end{pmatrix}$$

and here $a = 1.77$. Figure 3(*d*) is the superposition of two calculated reciprocal-quasilattice planes. One of them is obtained by introducing the phason strain described by M_2 with $a = 1.2$ and another is obtained by introducing M_3:

$$M_3 = \begin{pmatrix} (\tau - a)/(\tau a + 1) & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}.$$

The disappearance of some weak spots in figure 3(*b*) may be due to an intensity difference as discussed in section 3. A slight misfit seen from the shape of some spots may be due to some inhomogeneity of the phason strain. The agreement between figures 3(*d*) and 3(*b*) indicates that, at the onset of crystallization, a thermal phason strain is introduced so that the imperfect quasicrystal distorted further towards the crystalline state.

After the crystallization, only a few diffraction spots can be seen on the screen and they do not belong to a single net. This means that the degenerate fivefold axis of the quasicrystal is not parallel to any zone axis of the R phase exactly. When the degenerate fivefold axis deviates from the incident electron beam to some extent, it was found that at the beginning of crystallization the diffraction spots of the $[21\bar{1}]$ zone of the R phase (bottom left in figure 4) coexist with spots of the distorted icosahedral phase (top right in figure 4). The spots indicated by five triangular arrows in figure 4 are the same spots as shown in figure 3(*b*), and the corresponding reciprocal-quasilattice points are shown in figure 3(*d*), which are also indicated by triangular arrows but, because the $[21\bar{1}]$ zone axis of the R phase deviates from the incident beam in another direction, we can say only that the $[21\bar{1}]$ zone axis in the R phase is close to the fivefold axis in the icosahedral phase.

5. Conclusions

The rapidly solidified Al–Cu–Mg alloy examined has the structural features of both the icosahedral phase and the BCC phase and hence belongs to an intermediate state between

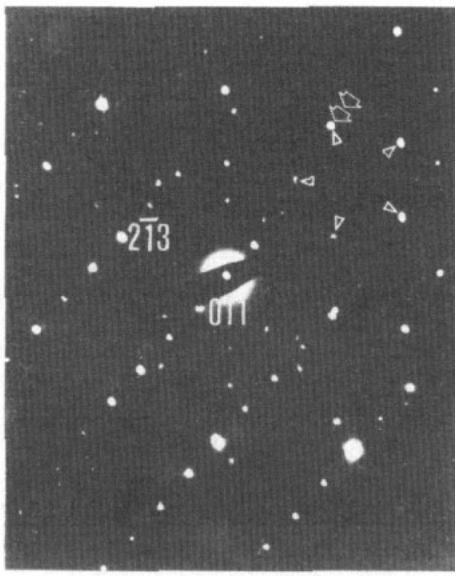

Figure 4. Electron diffraction pattern of rapidly solidified Al–Cu–Mg alloy taken during *in situ* heating at 284 °C (after figure 2(*b*), at the beginning of crystallization).

crystals and quasicrystals. The intermediate state may approximately be a two-dimensional quasicrystal. However, this two-dimensional quasicrystal is different from the decagonal phase; it is formed when the quasiperiodicity along one of the twofold axes changes into periodicity. The intermediate state can be interpreted as an imperfect icosahedral quasicrystal distorted by a linear phason strain. In addition, a thermal phason strain will be introduced into the quasicrystal when it is heated to the temperature of crystallization. We suggested that the phase transformation from imperfect quasicrystals to crystals is accompanied by a change in phason strain.

Acknowledgment

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