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Convergent-Beam and Small-Area-Parallel-Beam Electron Diffraction Patterns of Icosahedral Quasicrystals of an $Al_{74}Mn_{20}Si_6$ Alloy

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Abstract

Convergent-beam electron diffraction (CBED) and small-area-parallel-beam electron diffraction have revealed the crystallographic nature of icosahedral quasicrystals in a melt-quenched $Al_{74}Mn_{20}Si_6$ alloy. This quasicrystal possesses a much greater ordering in its atomic arrangement than an Al_6Mn quasicrystal. The present alloy shows little increase in the intensities of the reflections appearing at high scattering angles even when it is cooled to the temperature of liquid nitrogen, indicating its high Debye temperature. A careful examination has revealed that the point group of the present alloy is centrosymmetric $m\bar{3}\bar{5}$, although a small breakdown of mirror symmetry is observed. Small-area-parallel-beam electron diffraction patterns taken with the incidence direction along the fivefold axis showed characteristic displacements of the reflections from tenfold symmetry in spite of the greater ordering of the alloy. A tentative explanation of the diffraction patterns is given based on the knowledge obtained in normal crystallography.

1. Introduction

In a previous paper (Tanaka, Terauchi, Hiraga & Hirabayashi, 1985), we reported the crystallographic nature of the icosahedral quasicrystalline Al_6Mn alloy using convergent-beam electron diffraction (CBED) and small-area-parallel-beam electron diffraction. The quasicrystal has no fivefold, threefold or twofold axes and has no inversion center. It was found that the quasicrystals are highly strained.

It was reported that it is possible to prepare less strained quasicrystals (Chen & Chen, 1985) by adding several percent of silicon to the alloy. Bendersky & Kaufman (1986) prepared such a less strained quasicrystalline $Al_{71}Mn_{23}Si_6$ alloy and examined its point group using CBED. They obtained zone-axis CBED patterns which showed symmetries of $10mm$, $6mm$ and $2mm$ in the zeroth Laue-zone disks and $5m$, $3m$ and $2mm$ in higher-order Laue-zone (HOLZ) rings. From these results they identified the point group to be centrosymmetric $m\bar{3}\bar{5}$ (Hahn, 1983).

In the present study we have examined the crystallographic nature of a stabilized icosahedral quasicrystalline $Al_{74}Mn_{20}Si_6$ alloy using CBED and small-area-parallel-beam electron diffraction, and discuss its structural nature in comparison with that of Al_6Mn and $Al_{71}Mn_{23}Si_6$.

2. Experimental

Specimens of a melt-quenched $Al_{74}Mn_{20}Si_6$ alloy were supplied by Professor T. Masumoto and Dr A. Inoue of the Research Institute for Iron, Steel and Other Materials, Tohoku University. Specimens of $Al_{71}Mn_{23}Si_6$ were prepared by Drs Bendersky and Kaufman and supplied by Professor S. C. Moss, University of Houston, USA. The convergent-beam and parallel-beam electron diffraction studies of this material were performed with a JEM 100CX electron microscope equipped with a field-emission gun (FEG) and also a JEM 2000FX. The performance of the former instrument was described in a previous paper (Tanaka, Terauchi, Hiraga & Hirabayashi, 1985).

3. Results and discussion

Fig. 1(a) shows an electron micrograph obtained from an Al_6Mn alloy. It exhibits contrast on a scale of less than 10 nm, presumably owing to lattice strain, as already reported by many investigators. It is difficult to recognize definite grains of the quasicrystal. Figs. 1(b) and (c) show bright- and dark-field electron micrographs from an $Al_{74}Mn_{20}Si_6$ alloy. Quasicrystalline grains of dimensions $\sim$0·1 μm are seen as indicated by A, B, C, D.... Misorientation between neighbouring grains is a few millirad. Nishitani, Kawaura, Kobayashi & Shingu (1986) observed precipitates of a dodecahedron with protuberances, the whole having icosahedral symmetry, in a rapidly quenched and successively annealed Al-6·0 at.% Mn alloy. Chemical etching allowed us by chance to find that an assembly of grains, as seen in Fig. 1(b), resembled the precipitate shown in Fig. 2 of their paper. Thus it was found that the growth of such

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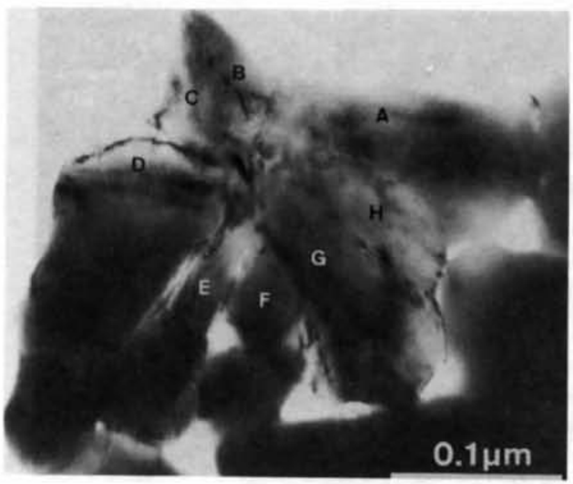
(a)



(b)

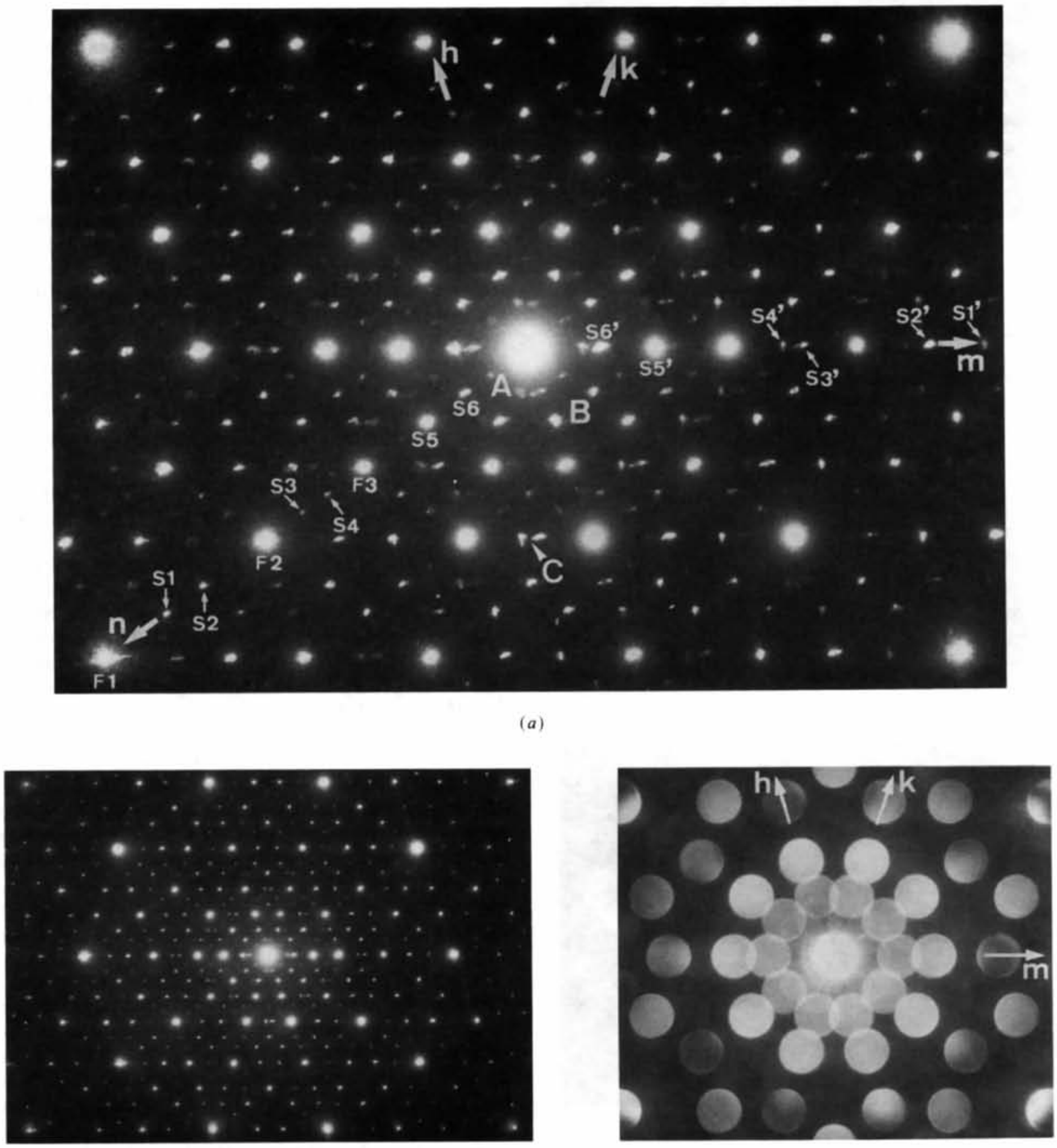
(c)

Fig. 1. (*a*) Electron micrograph of Al_6Mn shows contrast on a scale of less than 10 nm, presumably owing to lattice strain. (*b*) Bright- and (*c*) dark-field micrographs of $Al_{74}Mn_{20}Si_6$ show quasicrystalline grains of 0·1 µm dimensions and thickness contours.

precipitates occurs not only in the case of such special heat treatment as performed by Nishitani *et al.* (1986), but also in the present case. The grains show thickness contours and each appears to have mono-orientation. Electron-diffraction experiments, however, revealed that each grain consists of a number of subgrains less than 50 nm in diameter, which make a small but finite angle with each other (see below).

Fig. 2(*a*) shows a small-area-parallel-beam diffraction pattern taken from a 110 nm diameter area with an incident-beam divergence of 3×10^{-5} rad. Fig. 2(*b*) is a wide-angle pattern of Fig. 2(*a*). The reflections *B* of a lattice spacing of 1·37 nm have strong intensity; they were not observed in Al_6Mn (Tanaka, Terauchi, Hiraga & Hirabayashi, 1985) and were weakly observed in $Al_{71}Mn_{23}Si_6$ (Bendersky & Kaufman, 1986). The innermost reflections *A* are of a lattice spacing 2·27 nm, which were not observed even in $Al_{71}Mn_{23}Si_6$ (Bendersky & Kaufman, 1986). In the present pattern many weak diffraction spots appear, which were not observed in the two previous alloys, between strong reflections (Fig. 2*b*). Diffraction patterns taken with the electron incidences parallel to the twofold and threefold axes also display more spots than those from the previous alloys. Intensity measurement of all these reflections helps to construct a better model of the quasicrystal. The reflections at higher scattering angles (Fig. 2*b*) have stronger intensity than in Al_6Mn. These indicate that the present icosahedral phase has a much greater ordering. The quasicrystal may have more than three differently sized atomic sites and appears to be stabilized by the addition of a different-sized atom, Si. The reflections of this quasicrystal, consisting of many fine spots, show a larger and more inhomogeneous half-width than those of Al_6Mn for the same probe size. A typical reflection *C* in Fig. 2(*a*) shows an angular width of $1\cdot9\times10^{-4}$ rad in the vertical direction and $6\cdot4\times10^{-4}$ rad horizontally. This indicates that the misorientation between the subgrains is larger in the present case, and suggests that the greater ordering in a grain may produce the accumulation of strain at the grain boundaries. Examination of $Al_{71}Mn_{23}Si_6$ shows that the fine spots have a similar half-width but that the aggregation of the spots has a rather circular distribution about their center.

It was reported in a previous paper (Tanaka, Terauchi, Hiraga & Hirabayashi, 1985) that there is a zig-zag deviation of the three principal reflections from the radial line in a diffraction pattern taken with the electron incidence parallel to a fivefold axis. A halfway displacement of lattice fringes corresponding to the deviation is seen in a micrograph taken by Hiraga, Hirabayashi, Inoue & Masumoto (1985*a*,*b*), and is interpreted in terms of inhomogeneously quenched phason strains (Lubensky, Socolar, Steinhardt, Bancel & Heiney, 1986). In the present case, notwithstanding a much better ordering, such a deviation is still observed in the two directions denoted by h and k in Fig. 2(a). Fig. 2(c) shows a CBED pattern in which the disks are slightly overlapped to visualize the displacement of diffraction spots. It is clearly seen that the second disk in the direction h overlaps with that in the direction k to a greater extent than other disks in other directions, and the equivalent disk in the direction m, which is perpendicular to the direction midway between the directions h and k, radially overlaps



(a)

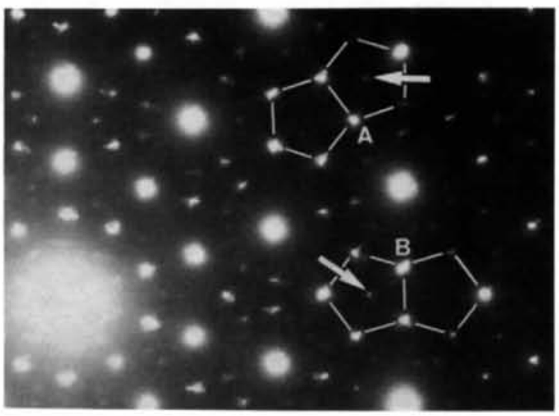
(b)

(c)

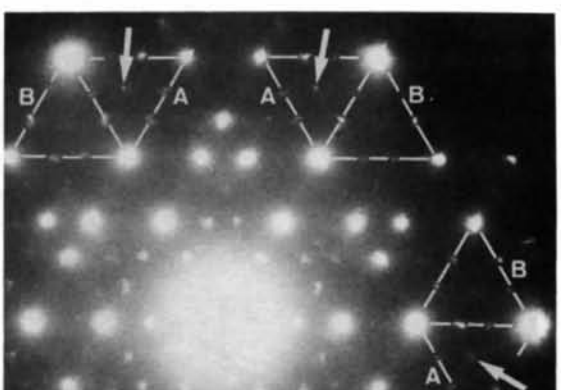
Fig. 2. (a) Small-area-parallel-beam diffraction pattern taken from a 110 nm diameter area of $Al_{74}Mn_{20}Si_6$ with an incident beam divergence of 3×10^{-5} rad. (b) Wide-angle pattern of (a) showing more spots than those of other quasicrystalline alloys. (c) CBED pattern revealing the deviations of positions of reflections.

more with the outer disk than in the other directions. The displacement is discussed in the last part of this paper.

Fig. 3(*a*) shows a part of a small-area-parallel-beam electron diffraction pattern taken from about 100 nm diameter area with the electron incidence parallel to a fivefold axis. This does not show a tenfold rotation symmetry, but a fivefold one with respect to the direct spot. That is, the two pentagons *A* drawn with white lines share one side. Only the outer pentagon contains a weak spot in its center. The pentagons *B* are equivalent with respect to the tenfold axis located at the direct beam. In a reverse manner to the case of the pentagons *A*, the inner pentagon of *B* contains a weak spot. These results prove the lack of a tenfold symmetry. We thought that these weak spots belonged to the zeroth Laue zone and were generated by *Umweganregung via* HOLZ reflections, and then thought that the quasicrystal was noncentrosymmetric



(*a*)



(*b*)

Fig. 3. Small-area-parallel-beam diffraction patterns of $Al_{74}Mn_{20}$-Si_6 taken with electron incidences parallel to fivefold (*a*) and threefold (*b*) axes, respectively. Note that the weak spots due to the tails of higher-order Laue-zone reflections appear in the pentagons and triangles.

(Tanaka, Terauchi, Hiraga & Hirabayashi, 1986). However, this was found to be incorrect. These weak reflections are the tails of profiles of the first-order Laue-zone reflections 322101 and 333101 in Elser's (1985) indexing system. Sometimes such weak reflections were observed at positions which are equivalent with respect to the tenfold axis, not to the fivefold axis. This fact indicates that quasicrystallites have two variants in their orientation, these being related to each other by 36 or 180° rotation about the axis. It was found that the whole of a precipitate has one variant. This means that precipitates which belong to the other variant also exist.

Fig. 3(*b*) shows a part of a small-area-parallel-beam diffraction pattern taken with the electron incidence parallel to a threefold axis. The weak spots of the first Laue zone shown with arrows have, not a sixfold, but a threefold rotation symmetry. Such weak reflections were sometimes observed at the positions equivalent with respect to the sixfold axis. That is, they were observed not only in the triangles *A*, but also in *B*, as in the case of Fig. 3(*a*). This result shows that there exists two variant quasicrystallites, these being related to each other by 60 or 180° rotation about the threefold axis. It was not possible, in spite of great care, to observe these weak first-Laue-zone reflections in $Al_{71}Mn_{23}Si_6$, presumably owing to the poor quality of this quasicrystal.

It is noted that the intensities of the reflections appearing at high scattering angles hardly increase, even when the specimen is cooled to the temperature of liquid nitrogen. This means that the Debye–Waller factor varies little between room temperature and liquid-nitrogen temperature and therefore that the quasicrystal has a high Debye temperature. An accurate measurement of the factor should be made.

Figs. 4(*a*)–(*f*) show three pairs of CBED patterns taken from an area about 10 nm thick and about 3 nm in diameter of an $Al_{74}Mn_{20}Si_6$ quasicrystal at an accelerating voltage of 60 kV. Each pair consists of a zeroth-Laue-zone pattern and a HOLZ pattern. The former pattern is produced approximately by the interaction of zeroth-Laue-zone reflections (two-dimensional or projection interaction). Distinct symmetries are seen in several disks, whereas none were observed in the disks from Al_6Mn. In the HOLZ patterns, many HOLZ rings and Kikuchi bands are clearly seen. The profiles of Kikuchi bands are symmetric with respect to their centers and are the same in the 5, 3 and 2 directions in Figs. 4(*b*), (*d*) and (*f*), respectively. These results contrast strongly with the Al_6Mn case, and indicate that the constituent atoms take a highly ordered arrangement in the quasicrystal.

The whole pattern (Buxton, Eades, Steeds & Rackham, 1976; Tanaka & Terauchi, 1985) formed with the zeroth-Laue-zone reflections (Fig. 4*a*), which shows the two-dimensional or projection symmetries, exhibits a tenfold rotation and two types of mirror

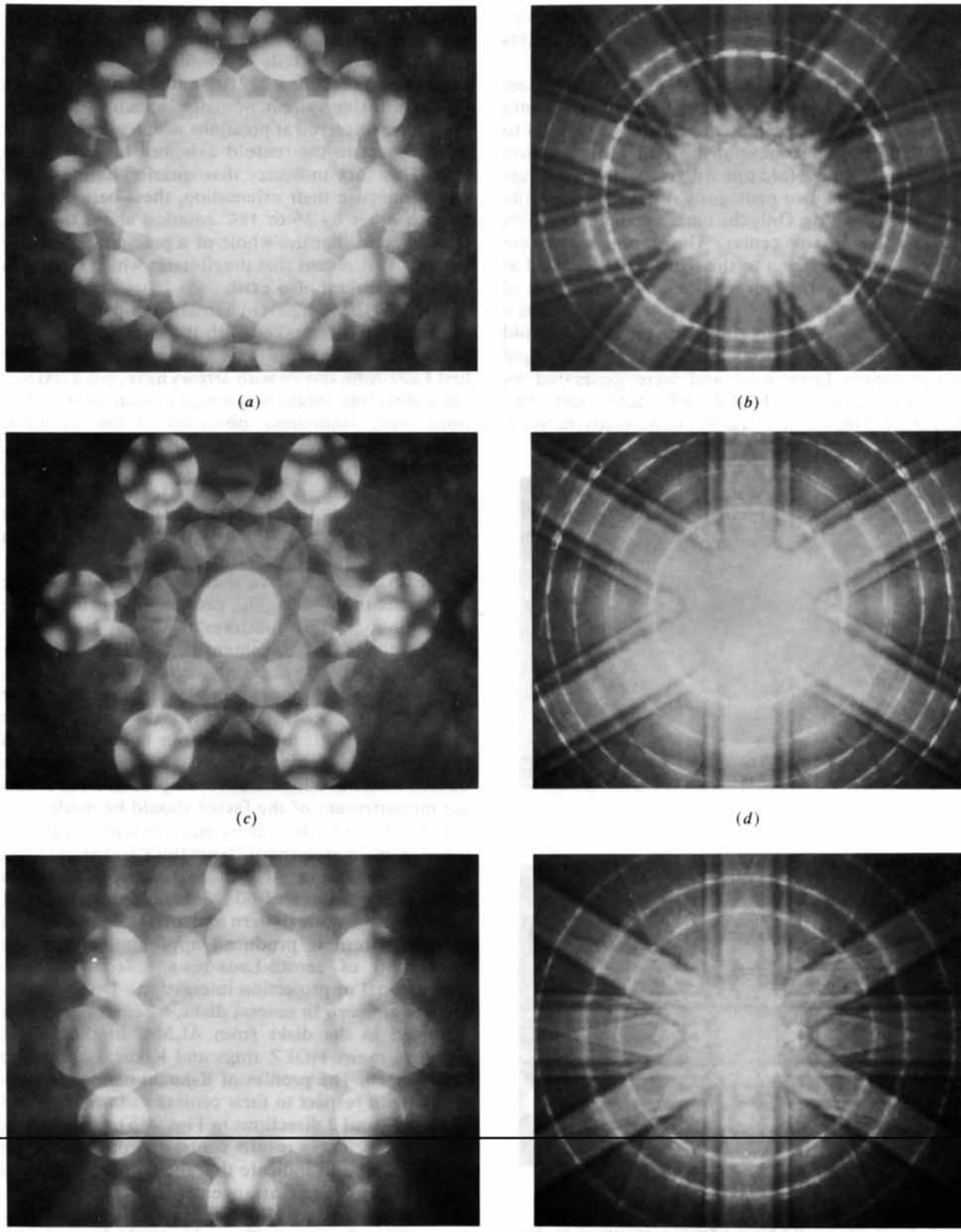


Fig. 4. Three pairs of zeroth-Laue-zone [(*a*), (*c*), (*e*)] and HOLZ [(*b*), (*d*), (*f*)] CBED patterns taken from an area about 10 nm thick and of about 3 nm diameter of $Al_{74}Mn_{20}Si_6$ at 60 kV. Symmetries are (*a*) 10*mm*, (*b*) 5*m*, (*c*) 6*mm*, (*d*) 3*m*, (*e*) 2*mm* and (*f*) 2*mm*.

Table 1. *Diffraction groups expected from the icosahedral point groups of* 235 *and* $m\bar{3}\bar{5}$ *for the incident beam parallel to the fivefold axis, and their symmetries appearing in the four types of CBED patterns*

Bright-field pattern (BP), whole pattern (WP), dark-field pattern (DP) and ±dark-field pattern (±DP) (Tanaka & Terauchi, 1985)

Point group	Diffraction group	BP	WP	DP	±DP
235	$5m_R$	$5m$	5	$\begin{cases}1\\ m_2\end{cases}$	$\begin{cases}1\\ m_R\end{cases}$ 1
	(Projection) $5m1_R$	$10mm$	$5m$	$\begin{cases}2=1_R\\ 2m_vm_2\end{cases}$	$\begin{cases}1\\ m_v1_R\end{cases}$ 1
$m\bar{3}\bar{5}$	10_Rmm_R	$10mm$	$5m$	$\begin{cases}1\\ m_2\\ m_v\end{cases}$	2_R 2_Rm_2 2_Rm_v
	(Projection) $10mm1_R$	$10mm$	$10mm$	$\begin{cases}2\\ 2m_vm_2\end{cases}$	21_R 21_Rm_v

symmetry, the resultant symmetry being expressed as $10mm$. The whole pattern formed with HOLZ reflections (Fig. 4*b*), which reveals the three-dimensional symmetries, is seen to have a fivefold symmetry and a type of mirror plane, the resultant symmetry being expressed as $5m$. Figs. 4(*c*) and (*d*) show symmetries $6mm$ and $3m$, respectively. Figs. 4(*e*) and (*f*) show the symmetry $2mm$.

It is known that there exist two icosahedral point groups, 235 and $m\bar{3}\bar{5}$ (Hahn, 1983). The former is non-centrosymmetric and has no mirror symmetry, but the latter is centrosymmetric. Table 1 shows the diffraction groups expected from these point groups at the incident beam parallel to the fivefold axis and their symmetries appearing in the four types of CBED patterns. Projection diffraction groups and their symmetries, in which only the interaction between zeroth-Laue-zone reflections is taken into account, are written in the second row. Diffraction groups obtained at other incident-beam directions are omitted, since these have been given in the literature (Buxton *et al.*, 1976; Tanaka & Terauchi, 1985). The whole-pattern symmetries observed in the present quasicrystal agree with those expected from point group $m\bar{3}\bar{5}$, as Bendersky & Kaufman (1986) obtained.

Fig. 5(*a*) shows a zone-axis CBED pattern taken with the electron incidence along the threefold axis. Figs. 5(*b*) and (*c*) show a pair of $\pm G$ dark-field CBED patterns. The pattern of the $+G$ dark-field disk coincides with that of the $-G$ disk by superposing the former on the latter with a translation vector of $-2G$. This symmetry indicates that the quasicrystal is centrosymmetric (Buxton *et al.*, 1976; Tanaka & Terauchi, 1985). Therefore, the quasicrystal was again confirmed to have point group $m\bar{3}\bar{5}$.

It is however noted that the symmetries investigated above are those appearing in intense reflections. To know the correct point group, the symmetries of weak reflections must be examined. Fig. 6 shows a CBED pattern taken from an area of about 3 nm diameter by tilting the specimen from a twofold axis into a direction in which a mirror symmetry is expected to exist. Weak reflection pairs AA' and BB', for instance, show a small breakdown of mirror symmetry. It is not clear at the present stage whether this breakdown has an important meaning for the identification of the point group or whether it can be attributed to residual strains of the specimen.

It is noted that a three-dimensional space-filling method with the Penrose-type skeleton was completed by Ogawa (1985). Hiraga & Hirabayashi (1986) proposed a structural model of a quasicrystal on the basis of Ogawa's method. This model appears to have a centrosymmetric atomic arrangement.

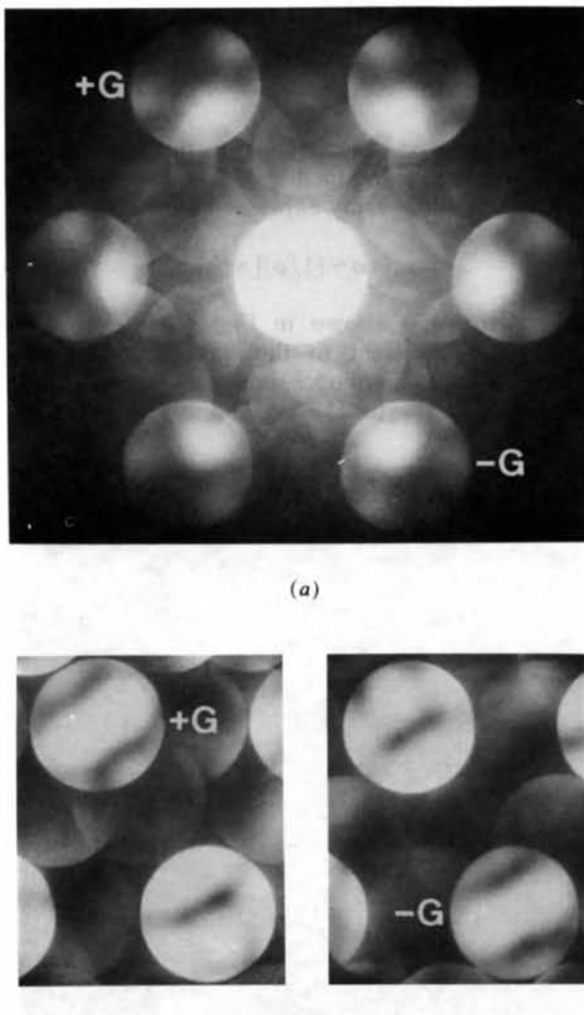


Fig. 5. CBED patterns of $Al_{74}Mn_{20}Si_6$ taken with the electron incidence along the threefold axis. (*a*) Zone-axis, (*b*) $+G$ dark-field and (*c*) $-G$ dark-field patterns. Translational symmetry appearing in the patterns (*b*) and (*c*) indicates the existence of an inversion center.

We return to discuss the diffraction pattern which shows the fivefold symmetry. In the above paragraphs, we have been concerned with the symmetry determination in assuming that the concept of the quasicrystal is realized. Close inspection of the diffraction patterns of Fig. 2(a), however, clarifies the following facts. All the spots in the directions m and n line up or show almost no angular deviation. These two directions are not equivalent, because the spots $S1$–$S6$ in the direction n are situated at different radial positions from the spots $S1'$–$S6'$ in the direction m. The diffraction spots in the other directions show angular deviations. The deviated positions of the reflections in these directions together with the non-deviated reflections can be produced by linear combinations of the reflection vectors in the directions m and n. Therefore, the two fundamental reciprocal-lattice vectors can be taken in the directions m and n for the diffraction pattern of Fig. 2(a) as in usual crystals, instead of requiring five fundamental reciprocal-lattice vectors for the two-dimensional quasilattice.

All the positions of the spots in the fundamental direction n are reproduced by the Fourier transform of a quasiperiodic array expressed by the equation

$$x_n = n + (1/\rho)[n/\tau]. \qquad (1)$$

This transform is shown in Fig. 7(a) for $\rho = \tau = (1+\sqrt{5})/2$, equivalent to the quasiperiodic array $LSLLSLSLL\ldots$ (Fibonacci sequence), where L and S have relative lengths $(1+\sqrt{5})/2$ and 1, respectively. The values of ρ and τ determine the ratio between the lengths of L and S, and the array of them, respectively. The positions of the peaks $F1$–$F3$ and $S1$–$S6$ correspond well to those of the reflections indicated with the same symbols in Fig. 2(a).

It is noted that the diffraction pattern of Fig. 7(a) resembles that of a long-period superlattice structure in a binary alloy which has an ordered array of two kinds of atoms and periodic antiphase domain boundaries (Johansson & Linde, 1936). That is, the strong reflections $F0$–$F3$ in Fig. 7(a) can be regarded as the fundamental reflections of the disordered (averaged) array, and the pairs of reflections ($S1$, $S2$), ($S3$, $S4$) and ($S5$, $S6$), symmetrically situated between the

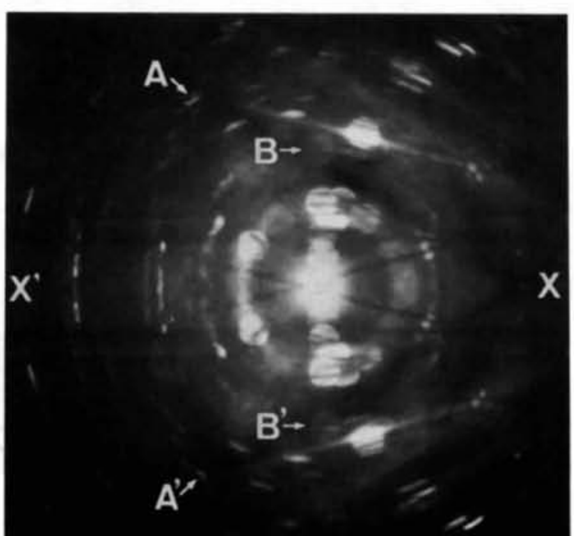


Fig. 6. CBED pattern of $Al_{74}Mn_{20}Si_6$ taken by tilting from a twofold axis shows a mirror symmetry about XX', although a small breakdown of the symmetry is observed between the reflection pairs AA' and BB'.

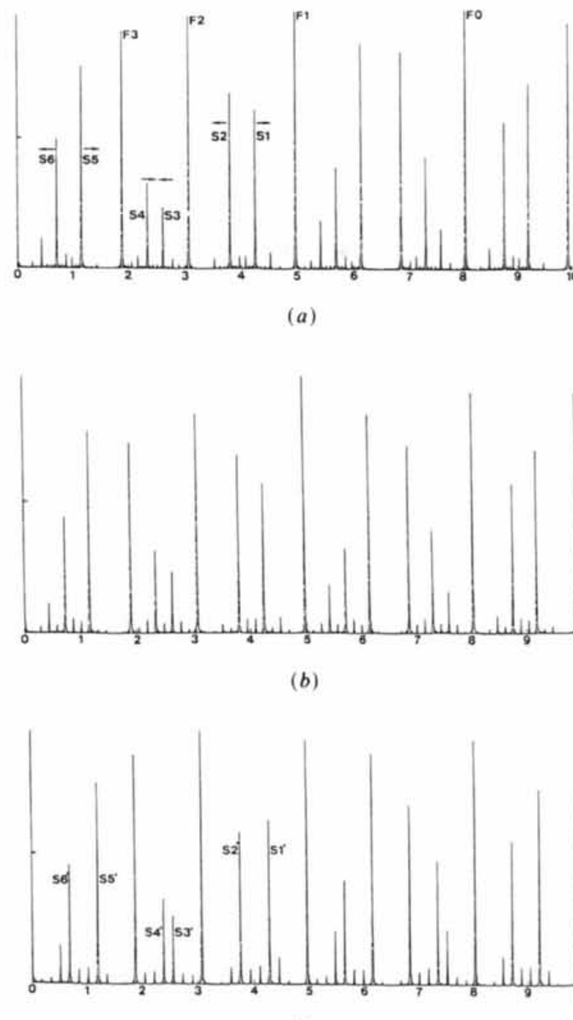


Fig. 7. Fourier transform of the quasiperiodic array generated by (a) the equation $x_n = n + (1/\rho)[n/\tau]$ for $\rho = \tau = (1+\sqrt{5})/2$, ($b$) that of the periodic array of $LSLLS$ as an approximation of the transform (a), and (c) that of the periodic array of $LLLS$ reproducing the coupled shifts of peaks indicated by arrows in (a). The abscissa shows $1/x_n$ and the ordinate shows intensity on an arbitrary scale.

fundamental reflections, can be regarded as the split superlattice reflections. In fact, the quasiperiodic array produced by (1) can be created to a good approximation by the introduction of the ordering of two elements (L and S) and periodic antiphase boundaries to the disordered array of L and S, although a uniform mixing of two long-period superlattice structures with different periods is needed to reproduce the incommensurate nature of the quasilattice (Fujiwara, 1957). Details of the method which creates the quasiperiodic array from the disordered array will be reported elsewhere.

The calculated diffraction pattern of a periodic array with a sequence *LSLLS* is shown in Fig. 7(b) as an approximation of the diffraction pattern of Fig. 7(a). These two patterns are very similar in position and intensity of the peaks, except for very weak peaks. This indicates that it is not easy to determine from the observed diffraction patterns whether the present alloy has a quasiperiodic or periodic array.

In contrast to the direction n, coupled shifts of the pairs of split superlattice reflections are seen along the fundamental direction m in Fig. 2(a). The shifts are shown schematically by arrows in the calculated pattern of Fig. 7(a). This suggests that there exists a change in the period of the antiphase boundaries in the language of the long-period superlattice in binary alloys. The sense of the shifts has been reproduced by a periodic array of *LLLS*, which cannot be created by (1), as shown in Fig. 7(c). (It is worthwhile to note that the opposite-sense shift to the present one is produced by a periodic array of *LS*.)

As a result, the experimental diffraction pattern of the present alloy (Fig. 2a) might be explained by such a crystal lattice which has tentative periodic arrays of *LSLLS* and *LLLS* in the directions n and m respectively, although the possibility of two-dimensional tiling with these arrays has not yet been investigated. Two-dimensional tiling using two motifs so as to create different periodic arrays in the two fundamental directions (and its extension to the three-dimensional case) must be studied. The fact that there exists such deviation from fivefold symmetry as seen in Fig. 2(a), even in the highly ordered quasicrystal of the present alloy, may indicate that the alloy does not form the quasicrystal in a precise meaning but rather forms such a crystal as described in this section.

4. Concluding remarks

Convergent-beam and parallel-beam electron diffraction using a brighter electron source (a field-emission gun) have been used successfully to investigate the crystallographic nature of quasicrystals. These methods have confirmed that the present quasicrystal belongs to the point group $m\bar{3}\bar{5}$, although a small breakdown of mirror symmetry exists. Further investigations are required to reveal whether the alloy of $Al_{74}Mn_{20}Si_6$ is correctly a quasicrystal or is still a crystal which consists of two motifs.

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