Transformation of the two-dimensional decagonal quasicrystal to one-dimensional quasicrystals: A phason strain analysis

H. Zhang and K. H. Kuo
Beijing Laboratory of Electron Microscopy, Academia Sinica, P.O. Box 2724, 100 080 Beijing, China, and Department of Materials Engineering, Dalian University of Science and Technology, 116 024 Dalian, China
(Received 7 September 1989)

The transformation of the two-dimensional (2D) decagonal quasicrystal to various 1D quasicrystals has been studied from the viewpoint of linear phason strain theory and compared with experiments. This theory accounts well for the shifts of electron-diffraction spots during this 2D-1D quasicrystalline transformation. In some cases five 1D quasicrystals can grow along the five twofold directions of the 2D decagonal quasicrystal as fivefold twins, and their composite electron-diffraction pattern has been simulated.

I. INTRODUCTION

After the first discovery of the icosahedrally related, two-dimensional (2D) decagonal quasicrystal in Al-Mn (Refs. 1 and 2) and Al-Fe (Ref. 3) alloys, many new ones have been found in other Al-*M* alloys, where *M* stands for transition metals including the platinum group metals.[4] This 2D decagonal quasicrystal displays tenfold rotational symmetry in a quasiperiodic plane and is periodic along the tenfold axis perpendicular to this aperiodic plane. If one of these two quasiperiodic directions becomes periodic, a 1D quasicrystal, i.e., periodic in two directions with the third one remaining aperiodic, will be resulted. This is indeed the case found recently by He *et al.*[5] in rapidly solidified Al-Ni-Si, Al-Cu-Mn, and Al-Cu-Co alloys. In addition to such 1D quasicrystals, artificial superlattices consisting of two different semiconductor[6] or metallic[7] layers with a thickness ratio close to the golden mean have also been synthesized.

Immediately after the discovery of 3D icosahedral and 2D decagonal quasicrystals, deviations from the perfect fivefold or tenfold symmetry were noted. The diffraction spots are either not lying on straight lines[8-10] or not arranged strictly in Fibonacci sequence.[9] The ten weak spots originally on a circle are now shifted to lie on an ellipse and in some cases the spots in the twofold direction become equally spaced. The latter was further proven by the equally spaced lines of image points in the high-resolution electron microscopic image.[9] Such a phenomenon was interpreted as the presence of local periodical translation[9,11] or linear phason strain.[10] As a matter of fact, they are somewhat equivalent if one introduces, as Elser[12] did, a flip-flop of two different tiles and considers this tiling mistake as a kind of phason strain.[13] For instance, the Fibonacci sequence of long (L) and short (S) intervals *LSLLSLSLLSLLS*. . . has a tiling mistake (underlined) or phason strain and becomes *LSLLSLSLLLSLS*. . . , then the three successive L's will form a local periodic translation order. However, the phason strain conception is theoretically more rigorous and practically more convenient to treat the spot-shift problem.[10]

Evidently, this kind of local periodic translation order can perhaps be considered as a kind of embryo of a crystalline phase in the icosahedral matrix.[9,11] Recently, Mai *et al.*[14,15] have treated the continuous quasicrystalline-to-crystalline transformation by the successive increase of linear phason strain in a quasicrystal. Following these works, we discuss the continuous transformation of the 2D decagonal quasicrystal into the 1D quasicrystal from the phason strain point of view and use simulated results to explain some observed experimental phenomena.

II. LINEAR PHASON STRAIN AND BRAGG PEAK SHIFT

Phonon and phason strains in a quasilattice have been discussed explicitly from the unit cell point of view by Socolor *et al.*[13] and this exposition of phason strain is rather convenient in handling the peak-shift problem of electron diffraction.[10,16] The readers are recommended to consult these papers and in the following we shall only give a brief introduction and the main conclusions.

In a quasicrystal there are more than one unit cell and in the case of 2D decagonal and 1D quasicrystals there are *two* unit cells arranged quasiperiodically forming a quasilattice. However, the density wave expression is still the same as that in a crystal:

$$\rho(\mathbf{r})=\sum_{\mathbf{G}}\rho_{\mathbf{G}}e^{i(\mathbf{G}\cdot\mathbf{r}+\phi_{\mathbf{G}})}\,, \tag{1}$$

except that the phase term $\phi_{\mathbf{G}}$ is now given by

$$\phi_{\mathbf{G}}=\mathbf{u}(\mathbf{r})\cdot\mathbf{G}^{\parallel}+\mathbf{w}(\mathbf{r})\cdot\mathbf{G}^{\perp}\ . \tag{2}$$

A linear phonon strain $\mathbf{u}$, as in a crystal, only gives rise to a pure translation of the quasilattice or a distortion of the unit cells but it will not change the configuration of the unit-cell arrangement. On the contrary, a linear phason strain $\mathbf{w}$ will produce a rearrangement of unit cells but no distortion or uniform translation of them. This is called phason simply because mathematically the $\mathbf{w}$ variable is analogous to the phason degree of freedom in an incommensurate modulated crystal when this is treated in a high-dimensional superspace.

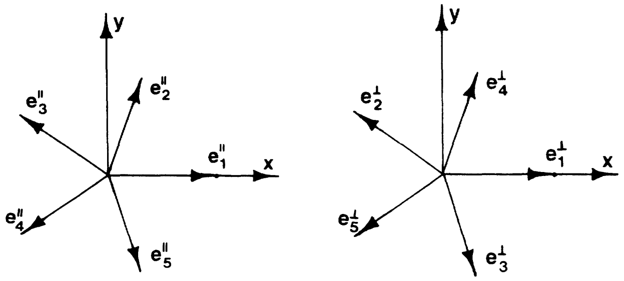

FIG. 1. Reciprocal vector bases of the 2D decagonal quasicrystal.

The $\mathbf{u}\cdot\mathbf{G}^{\parallel}$ term also occurs in a crystal and we call the $\mathbf{G}^{\parallel}$ a reciprocal vector in the real or physical space, whereas $\mathbf{G}^{\perp}$ that in the complementary, perpendicular, or pseudospace. In other words, in a high-dimensional superspace,

$$\mathbf{G}=\mathbf{G}^{\parallel}+\mathbf{G}^{\perp} . \tag{3}$$

The 2D decagonal quasilattice or the Penrose pattern can best be described by the five vectors directed toward the vertices of a pentagon in a 2D plane,[13] and the corresponding reciprocal bases are shown in Fig. 1. They are

$$\mathbf{G}^{\parallel}=\sum_{i=1}^{5} n_i \mathbf{e}_i^{\parallel}, \quad \mathbf{e}_i^{\parallel}=\{\cos(2j\pi/5),\ \sin(2j\pi/5)\} ,$$
$$j=i-1 \tag{4}$$

$$\mathbf{G}^{\perp}=\sum_{i=1}^{5} n_i \mathbf{e}_i^{\perp}, \quad \mathbf{e}_i^{\perp}=\{\cos(4j\pi)/5,\ \sin(4j\pi/5)\} ,$$
$$j=i-1 . \tag{5}$$

If the phason strain is approximately linear in a small region of a quasicrystal, it can be written as

$$\mathbf{w}(\mathbf{r})=\mathbf{w}(0)+\mathbf{r}\cdot\mathbf{M} , \tag{6}$$

where

$$\mathbf{M}=\begin{bmatrix} \underline{a} & \underline{b} \\ \underline{c} & \underline{d} \end{bmatrix} \tag{7}$$

is a second-rank tensor and $\mathbf{M}\cdot\mathbf{G}^{\perp}$ gives the shift of a Bragg peak. Then by the action of this phason strain the diffraction spot will occur at the end of $\mathbf{G}^{\parallel\prime}$:

$$\mathbf{G}^{\parallel\prime}=\mathbf{G}^{\parallel}+\mathbf{M}\cdot\mathbf{G}^{\perp} . \tag{8}$$

Obviously the shift of spot is controlled by the vector $\mathbf{G}^{\perp}$:[10] (i) Shifts in spots will be along the phason strain direction; (ii) the magnitude of the shifts will be proportional to $|\mathbf{G}^{\perp}|$; (iii) since the intensity of a spot decreases roughly with increasing $|\mathbf{G}^{\perp}|$, the faintest spots will be shifted the most. If there is also nonlinear phason strain, there will be spot broadening as well in the strain direction.

III. RESULTS AND DISCUSSION

The 2D decagonal quasicrystal in the Al-Cu-Co alloy is rather perfect as evidenced by the large number of sharp diffraction spots in the tenfold electron-diffraction pattern (EDP) shown in Fig. 2(a). This was further proven by the ten symmetrical Kikuchi bands in the convergent beam EDP.[5] Figure 2(b) is the simulated diffraction pattern of the 2D decagonal quasilattice (Penrose pattern) by the cut-and-projection method. The match between Figs. 2(a) and 2(b) is reasonably good, especially the concentric sets of decagons of weak spots marked with arrows. The spots along one of the ten twofold P directions in Fig. 2(b) is reproduced in line diagrams in Figs. 5(a) and 6(a) and it is clear that the intervals between two neighboring spots are in the ratio of $1:\tau:\tau^2$ obeying the Fibonacci τ relationship.

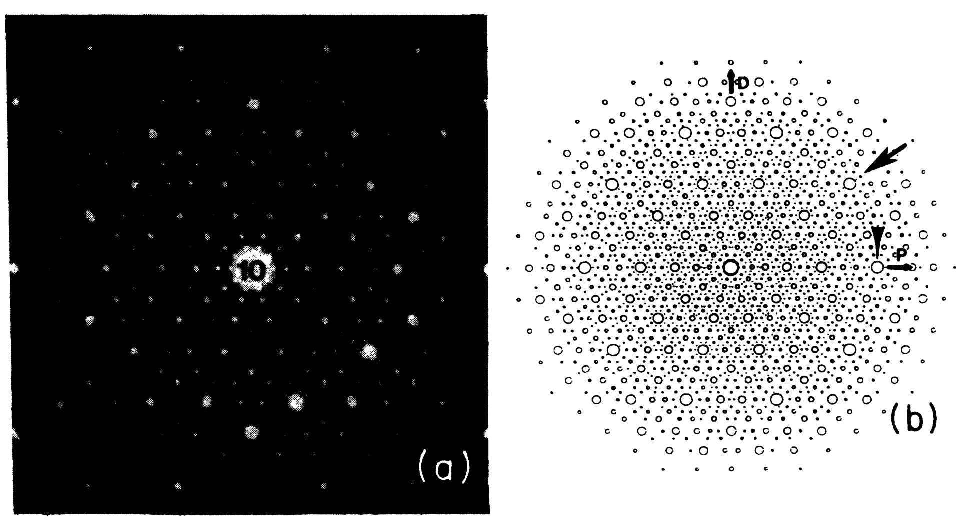

FIG. 2. (a) Experimental and (b) simulated tenfold electron-diffraction patterns of the 2D decagonal quasicrystal. The ten weak spots lying on a circle around a strong spot are marked with an arrow.

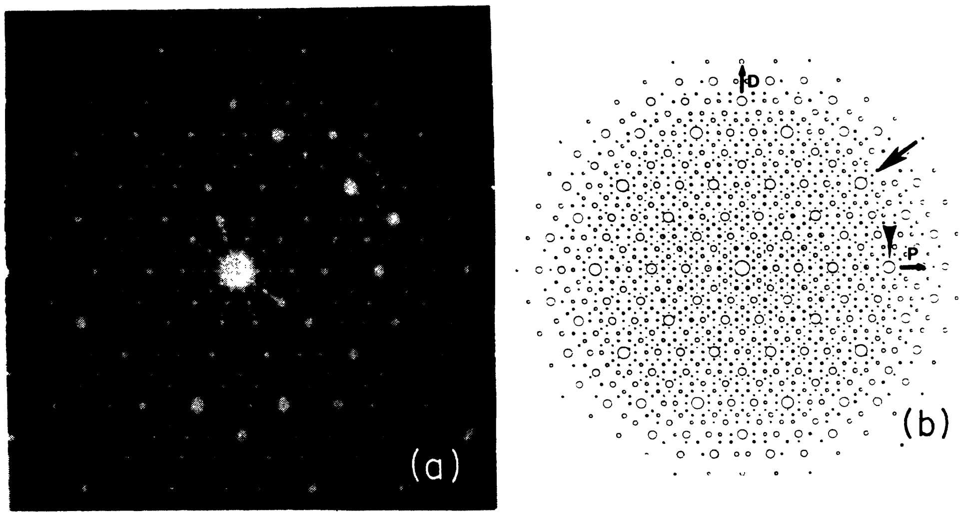

FIG. 3. (a) Experimental and (b) simulated pseudotenfold electron-diffraction patterns of the 1D quasicrystal. The ten weak spots lying on a circle in Fig. 2 are now on an ellipse, also marked with an arrow. The spots lying in the horizontal P direction are periodic giving a periodicity of $13\times0.3=3.9$ nm [there are thirteen spots between the central beam and the spot marked with an arrowhead corresponding to $(0.3\text{ nm})^{-1}$]. This line of spots is shown in Figs. 5(a) and 6(a) in line diagrams.

In some of the specimens heated to 800 °C for 40 h, the ten weak spots in the tenfold EDP are shifted to lie on an ellipse with the long axis in the D direction. They are also marked with an arrow in Fig. 3(a) in order to be compared with their counterparts in Figs. 2(a) and 2(b). Moreover, the spots along the horizontal twofold P direction now become equally spaced and there are altogether thirteen spots between the central transmitted beam and the strong spot marked with an arrowhead in Fig. 3. Tilting 90° brings the pseudotenfold axis down to the plane of Fig. 4 and lying in the vertical direction. This pattern is quite similar to the D EDP of the 2D decagonal quasicrystal with the six very strong spots forming a hexagon. In this case the equally spaced spots in the P direction become more evident. The strong spot marked with an arrowhead is $(0.3\text{ nm})^{-1}$ from the center and this shows the periodicity in the P direction in the real space being $13\times0.3=3.9$ nm. Figure 4 shows clearly the 2D periodicity and proves that the quasicrystal in question is a 1D one. In addition to this 1D quasicrystal, He *et al.*[5] have also reported other 1D quasicrystals with a periodicity of $3\times0.3=0.9$ and $5\times0.3=1.5$ nm, respectively. They suggested that these periodicities are the various approximants of the Fibonacci series $(1,2,3,5,8,13,\ldots)$

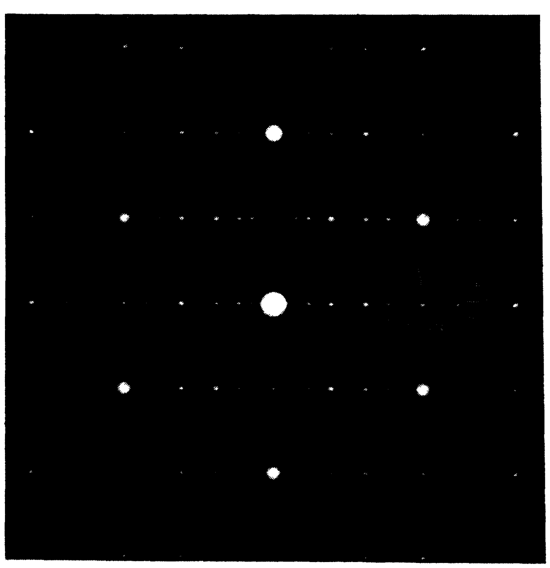

FIG. 4. The twofold D electron-diffraction pattern of the same 1D quasicrystal as in Fig. 3, showing clearly its 2D periodicity (Ref. 5).

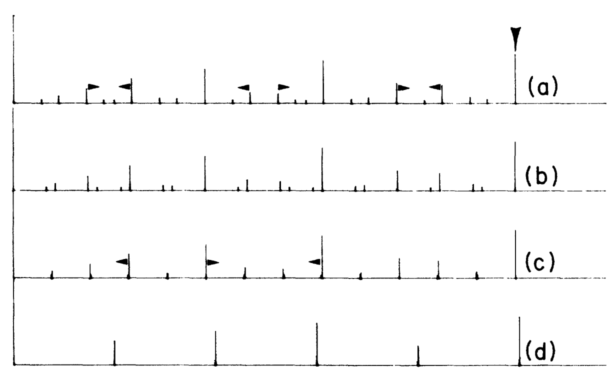

FIG. 5. Calculated positions and intensities of diffraction spots along the P direction, the element $\underline{a}$ of the phason strain tensor having a negative sign. (a) 2D decagonal quasicrystal with $\underline{a}=0$. Small arrowheads show the shift directions of spots in (b)–(d). (b) $\underline{a}=-0.01$; shifts in weak spot become obvious. (c) 1D quasicrystal with $\underline{a}=-0.02$; medium-to-strong spots start to move, becoming periodic and thirteen in number between the central beam to the strong spot marked with a big arrowhead. (d) 1D quasicrystal with $\underline{a}=-0.145$; only five strong spots appear periodically within this range.

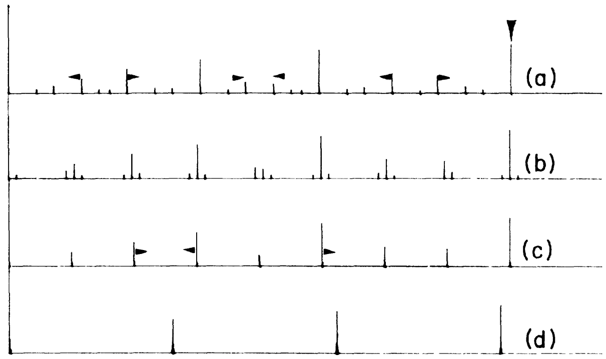

FIG. 6. Same as Fig. 5 except $\underline{a}$ is positive. (a) 2D decagonal quasicrystal with $\underline{a}=0$. The shift directions of strong spots are reversed comparing with Fig. 5; (b) $\underline{a}=0.04$, spot shifts are obvious; (c) 1D quasicrystal with $\underline{a}=0.055$, spots become periodic and are eight in number within this range; (d) 1D quasicrystal with $\underline{a}=0.38$, only three strong spots appear periodically within this range.

and that these 1D quasicrystals are transformed from the 2D decagonal quasicrystal.

In order to verify and follow the continuous transformation from the 2D decagonal quasicrystal to various 1D quasicrystals, we have introduced gradually the phason strain into the 2D decagonal quasicrystal. Looking at Fig. 3(a) carefully, all weak spots, except those in the vertical D direction, are shifted somewhat in the horizontal direction, but the shifts are not significant for medium to strong spots. This implies that the shifts in spots agree with the existence of a rather weak phason strain in this direction. The shifted spots are symmetrical with respect to the vertical D direction, which seems to be a mirror. This requires that the second-rank tensor **M** be symmetrical too.

First we chose the $\underline{a}$ element of **M** to be -0.01, all other elements being zero, and the distribution of spots along the P direction shown in Fig. 5(b) was obtained. Comparing with the corresponding 2D decagonal case shown in Fig. 5(a), the directions of movement of some of the spots are arrowed in Fig. 5(a). With $\underline{a}$ being further increased to -0.02, the simulated pseudotenfold pattern is shown in Fig. 3(b) and the distribution of spots in the twofold P direction in Fig. 5(c). Now the spots in the latter are equally spaced and are thirteen in number from the center to that marked with a big arrowhead. The ten weak spots marked with an arrow in Fig. 3(b) are on an ellipse almost identical to those marked also with an arrow in Fig. 3(a). The ten strong spots are more or less still lying on concentric circles in agreement not only with the experimental observation but also with the phason strain theory. With further increase of the phason strain the shifts of spots proceed further along the directions shown in Fig. 5(a) until $\underline{a}=-0.145$, then another 1D quasicrystal with a periodicity of $5\times0.3=1.5$ nm in the P direction is obtained, as illustrated in Fig. 5(d). Now only five equally spaced strong spots remain in the P direction; all weak spots either coincide with these strong spots or disappear in the background.

In order to simulate the spot distribution in the 1D quasicrystal shown in Fig. 6(c), the sign of $\underline{a}$ should be reversed, since now the strong spots in Fig. 6(c) are lying in the reverse directions of the corresponding spots in Fig. 5(b) compared with those in Figs. 5(a) or 6(a). This is verified by using $\underline{a}=0.04$ as shown in Fig. 6(b) and again the directions of spot shifts are marked with arrows in Fig. 6(a), which are in the opposite direction as those shown in Fig. 5(a). With further increase of $\underline{a}=0.055$, Figs. 6(c) and 7(a) are obtained. The latter is the simulated pseudotenfold pattern of another 1D quasicrystal with a periodicity of $8\times0.3=2.4$ nm along the P direction. This 1D quasicrystal has not been found yet, but it belongs to the missing link among the 1D quasicrystals with periodicities of 13×0.3, 5×0.3, and 3×0.3 nm in the P directions found earlier.[5] The ten weak spots in Fig. 7(a) shown by an arrow again lie on an ellipse similar to those shown in Fig. 3, but the shifts in spots in the present case are more obvious than in Fig. 3 and it can be seen easily through the arrowed twofold direction that the spots are not lying on a straight line. However, since

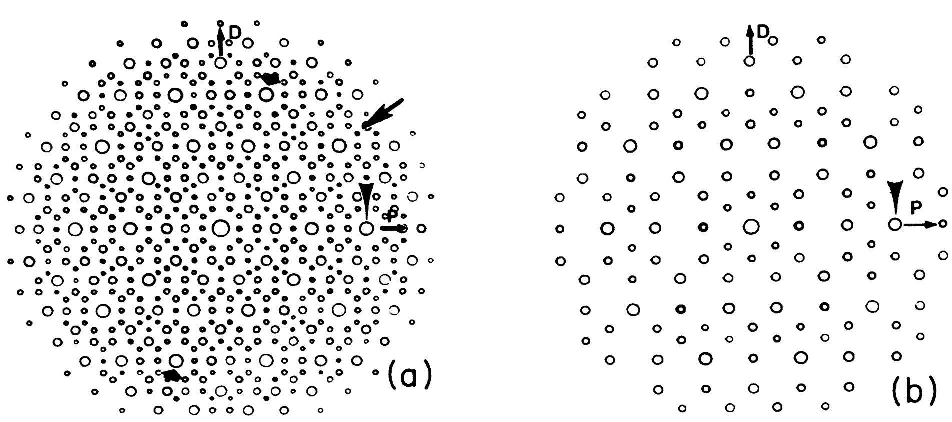

FIG. 7. Simulated pseudotenfold electron-diffraction patterns of two 1D quasicrystals. (a) $\underline{a}=0.055$, periodicity along P is 8×0.3 nm. The ten weak spots lying on an ellipse still can be observed. The spots are lying on horizontal "layer lines" following the Fibonacci sequence. (b) $\underline{a}=0.38$, periodicity along P is 3×0.3 nm. Only the strong spots are left.

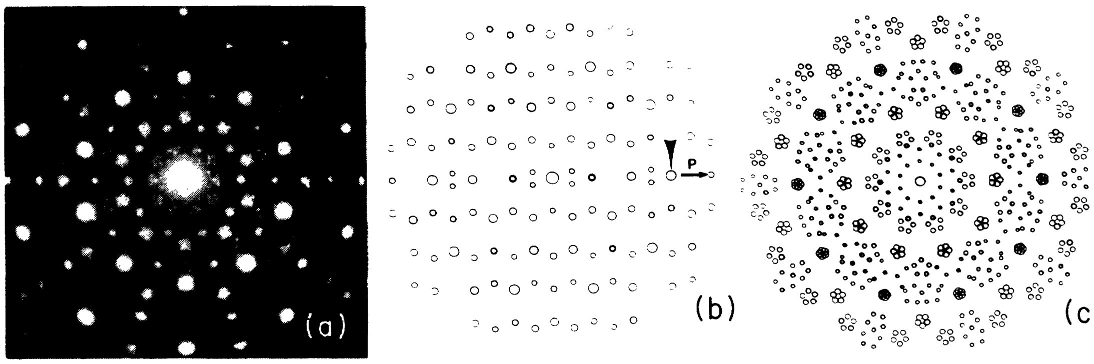

FIG. 8. (a) Tenfold electron-diffraction pattern of the Al-Ni-Si 2D decagonal quasicrystal. Each group of spots consists of five spots forming a small pentagon (Ref. 17). (b) Simulated pattern of 1D quasicrystal with $\underline{a}=\underline{d}=0.38$, $\underline{b}=\underline{c}=0$. Only strong spots are left. (c) Five such patterns superposed at an angle of 72° between two patterns. The composite pattern matches the observed electron-diffraction pattern in (a) fairly well, implying that the latter is a composite pattern of five differently oriented 1D quasicrystals.

the shift of spot is in the horizontal direction, all spots in Fig. 7(a) are still lying on a set of horizontal "layer lines" following the Fibonacci sequence in the D direction. This exhibits the 1D quasiperiodic translational order.

Further increasing the phason strain to $\underline{a}=0.38$ results in the 1D quasicrystal with a periodicity of $3\times0.3=0.9$ nm, as shown by the simulated patterns in Figs. 6(d) and 7(b). The ellipse consisting of weak spots no longer exists, and neither do all the other weak spots. Now even the sets of ten strong spots are lying on an ellipse in Fig. 7(b), implying that the presence of severe phason strains produces significant shifts of all spots, including the strong ones. The number of spots is also reduced materially and their quasiperiodic "layer lines" characteristics become more obvious.

Thus we have simulated the pseudotenfold EDP's of all the observed 1D quasicrystals by successively increasing the magnitude of only one element $\underline{a}$ of the phason strain tensor. However, we need to have both positive and negative signs of this element to recover the EDP's of this series of 1D quasicrystals. These are the more ideal cases and reality is more complex. For instance, Fig. 8(a) is a tenfold EDP frequently encountered in the Al-Ni-Si quasicrystal[17] and many "spots" with a pentagonal shape are in fact groups of five spots forming small pentagons. This indicates that this EDP may be a composite one resulting from five superposed EDP's, as it has been found and explained before in the distorted tenfold EDP's of an icosahedral quasicrystal.[10,11] This was experimentally proven by using microdiffraction with a focused beam.[11] In a small region of several tens of nanometers, only one variant or two differently oriented variants of this 1D quasicrystal existed and the EDP looked more or less similar to that shown in Fig. 7(b). The periodicity in the twofold P direction is the same but the shifts of other spots are more pronounced. After several trials, Fig. 8(b) was obtained, which is almost identical to the microdiffraction pattern and in this case both the diagonal tensor elements $\underline{a}$ and $\underline{d}$ are 0.38. A superposition of five such simulated patterns with an angle of 72° between the two neighboring ones is shown in Fig. 8(c). The match between Figs. 8(a) and 8(c) is good enough to show that Fig. 8(a) is a composite EDP of five twins of the 1D quasicrystals transformed from the 2D decagonal quasicrystal along its five twofold P directions. This may also serve to prove the success of applying linear phason strain to account for the shifts in spots in an EDP and to follow the continuous transformation of 2D quasicrystal into a 1D one.

From the simulation and discussion presented above it becomes clear that the 1D quasicrystal is only an intermediate stage of the continuous transformation of the 2D decagonal quasicrystal to a related crystalline phase. The 1D quasicrystal must have a certain metastability so that EDP's of it have been frequently observed. However, just as it has been proven that the Al-Mn 3D icosahedral quasicrystal can transform either directly to a related crystalline phase or through the 2D decagonal quasicrystal as an intermediate state,[18] the 2D decagonal quasicrystal can also transform either directly to a related crystalline phase or through the 1D quasicrystal as a metastable intermediate state. In any case, the possibility of such a direct transformation should exist and we have already found some evidence of such a direct transformation. This and the 1D quasicrystalline-to-crystalline transformation are now under investigation.

ACKNOWLEDGMENTS

The authors thank L. X. He for providing the EDP's of the Al-Cu-Co 1D quasicrystal and X. Z. Li for Fig. 8(a).

[1]L. Bendersky, Phys. Rev. Lett. **55**, 1461 (1985).
[2]K. Chattopadhyay, S. Lele, S. Ranganahan, G. N. Subbana, and N. Thangaraj, Current Sci. **54**, 895 (1985).
[3]K. K. Fung, C. Y. Yang, Y. Q. Zhou, J. G. Zhao, W. S. Zhan, and B. G. Shen, Phys. Rev. Lett. **56**, 2060 (1986).
[4]K. H. Kuo, Mater. Sci. Forum **22-24**, 131 (1987).
[5]L. X. He, X. Z. Li, Z. Zhang, and K. H. Kuo, Phys. Rev. Lett. **61**, 1116 (1988).
[6]R. Merlin, K. Bajema, R. Clarke, F. Y. Juang, and P. K. Bhattacharya, Phys. Rev. Lett. **55**, 1786 (1985).
[7]A. Hu, C. Tien, X. J. Li, Y. H. Wang, and D. Feng, Phys. Lett. **119A**, 313 (1986).
[8]M. Tanaka, M. Terauchi, K. Hiraga, and M. Hirabayashi, Ultramicrosc. **17**, 279 (1985).
[9]K. H. Kuo, J. Phys. (Paris) Colloq. **47**, C3-425 (1986).
[10]P. A. Bancel and P. A. Heiney, J. Phys. (Paris) Colloq. **47**, C3-341 (1986).
[11]Z. Zhang and K. H. Kuo, J. Microsc. **146**, 313 (1987).
[12]V. Elser, Phys. Rev. Lett. **54**, 1730 (1985).
[13]J. E. S. Socolor, T. C. Lubensky, and P. J. Steinhardt, Phys. Rev. B **34**, 3345 (1986).
[14]Z. H. Mai, S. Z. Tao, B. S. Zhang, and L. Z. Zeng, Phys. Rev. B **38**, 1291 (1988).
[15]Z. H. Mai, L. Xu, N. Wang, K. H. Kuo, Z. C. Jin, and G. Cheng, Phys. Rev. B **40**, 12 183 (1989).
[16]T. C. Lubensky, J. E. S. Socolar, P. J. Steinhardt, P. A. Bancel, and P. A. Heiney, Phys. Rev. Lett. **57**, 1440 (1986); J. E. S. Socolor and D. C. Wright, *ibid.* **59**, 221 (1987).
[17]X. Z. Li and K. H. Kuo, unpublished.
[18]R. J. Schaefer and L. Bendersky, Scr. Metall. **20**, 745 (1986).

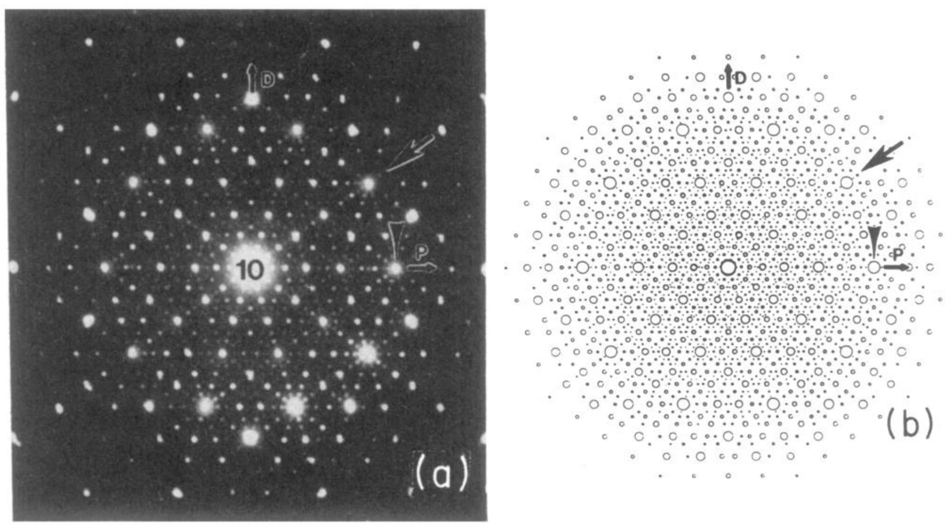

FIG. 2. (a) Experimental and (b) simulated tenfold electron-diffraction patterns of the 2D decagonal quasicrystal. The ten weak spots lying on a circle around a strong spot are marked with an arrow.

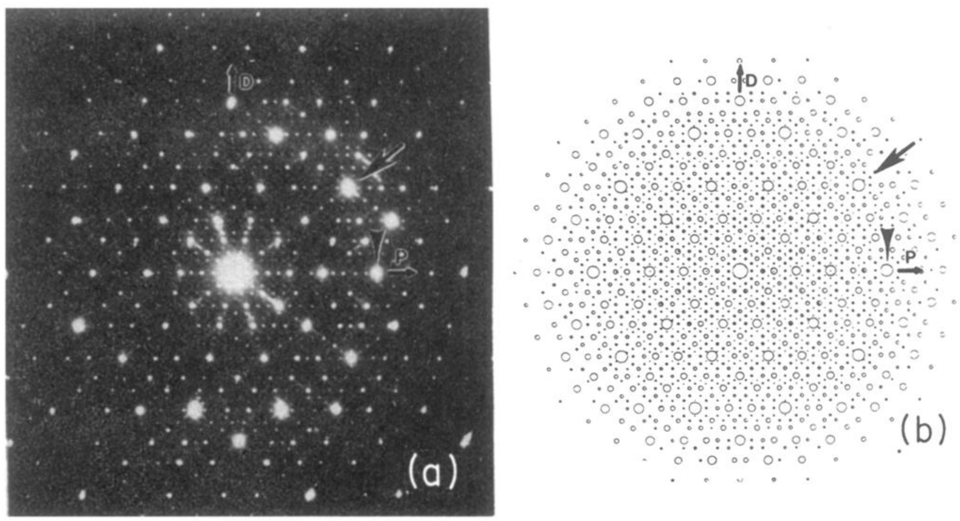

FIG. 3. (a) Experimental and (b) simulated pseudotenfold electron-diffraction patterns of the 1D quasicrystal. The ten weak spots lying on a circle in Fig. 2 are now on an ellipse, also marked with an arrow. The spots lying in the horizontal P direction are periodic giving a periodicity of $13\times0.3=3.9$ nm [there are thirteen spots between the central beam and the spot marked with an arrowhead corresponding to $(0.3\text{ nm})^{-1}$]. This line of spots is shown in Figs. 5(a) and 6(a) in line diagrams.

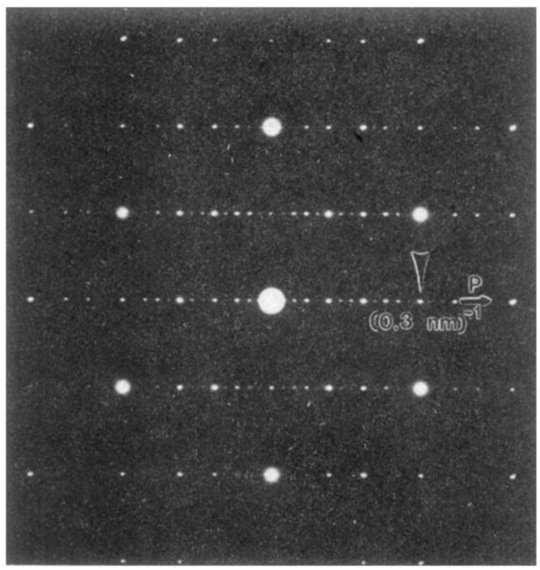

FIG. 4. The twofold D electron-diffraction pattern of the same 1D quasicrystal as in Fig. 3, showing clearly its 2D periodicity (Ref. 5).

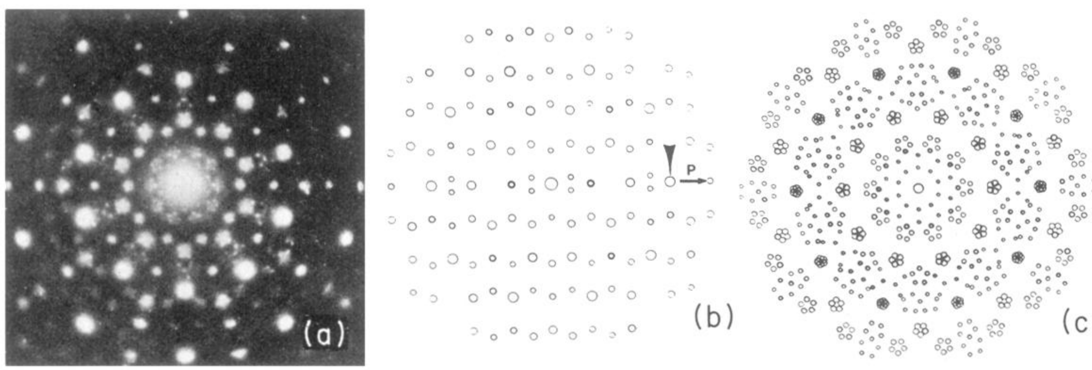

FIG. 8. (a) Tenfold electron-diffraction pattern of the Al-Ni-Si 2D decagonal quasicrystal. Each group of spots consists of five spots forming a small pentagon (Ref. 17). (b) Simulated pattern of 1D quasicrystal with $\underline{a}=\underline{d}=0.38$, $\underline{b}=\underline{c}=0$. Only strong spots are left. (c) Five such patterns superposed at an angle of 72° between two patterns. The composite pattern matches the observed electron-diffraction pattern in (a) fairly well, implying that the latter is a composite pattern of five differently oriented 1D quasicrystals.