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A continuous transition from twins to ‘quasi-crystals’

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Abstract. In this paper we discuss different structures, built up using two types of Robinson triangle tiling the plane without defects and having diffraction patterns with symmetry forbidden by classical crystallography. A universal behaviour of the diffraction patterns of the different structures allows definition of a similarity parameter which distinguishes ‘quasi-crystals’ (i.e. structures for which the diffraction peak intensities scale as N^2) from all other types of structure. It is shown that the similarity parameter depends only on the concentration of small Robinson triangles. The concentration dependence of the similarity parameter exhibits singular behaviour at the Penrose concentration, indicating that a continuous transition takes place from twinned structures to ‘quasi-crystals’. The critical exponent for this behaviour was determined to be 0.55 ± 0.02 .

1. Introduction

The experimental observation of systems showing diffraction patterns which exhibit symmetry forbidden by the concepts of classical crystallography (Shechtman *et al* 1984) has been theoretically explained by many different models (Penrose 1974, Mackay 1982, Sadoc and Mossieri 1982, Duneau and Katz 1985, Kramer 1985, Pauling 1985, 1987, Bak 1986, Wolny and Lebech 1986, Fruchart and Dubois 1987, Janot *et al* 1987, Jønsson and Andersen 1988). Among these models there are two concepts: Penrose-like structures (Penrose 1974, Mackay 1982) and crystal twinning (Pauling 1985, 1987, Fruchart and Dubois 1987), which have been used to obtain diffraction patterns with fivefold symmetry. Recently the concept of random quasi-crystalline structures obtained by the growth method, i.e. by attaching tiles to an existing seed (Stephens and Goldman 1986, Minchau *et al* 1987, Nori *et al* 1988, Ronchetti *et al* 1988, Wolny *et al* 1988, Lebech *et al* 1988, Onada *et al* 1988), or equilibrated by the Monte Carlo method (Widom *et al* 1987, Strandburg *et al* 1989) or by molecular dynamics (Lancon and Billard 1988) has been extensively studied. Theoretical descriptions for such structures using transfer matrix calculations (Henley 1988, Widom *et al* 1989) have also been presented. Calculations of the diffraction patterns using the analysis of the two-dimensional hypersurface projected from the five-dimensional hyperspace to the physical space can be also found (Jarić 1986, Elser 1985, 1986, Henley 1988).

In this paper we compare different types of two-dimensional structure, which can explain the observed fivefold or tenfold symmetry of calculated diffraction patterns. We have studied several different structures which can be obtained by tiling the plane without defects using only two types of Robinson triangle (Robinson 1975, Godreche

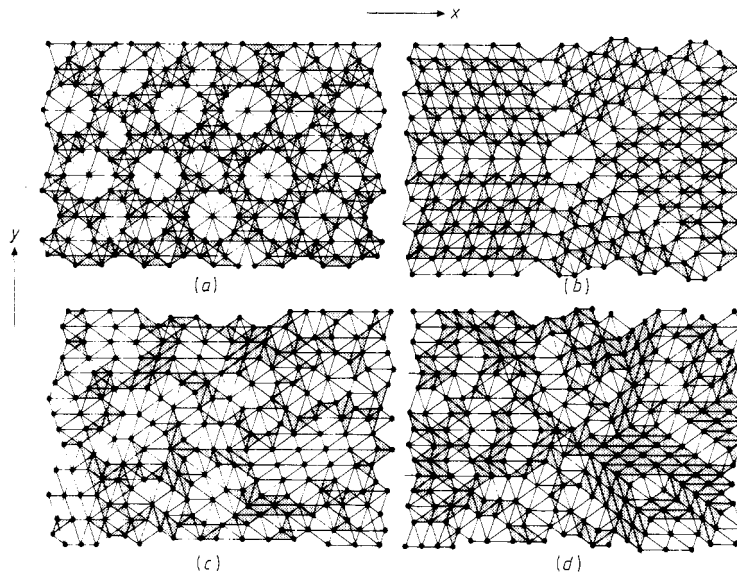


Figure 1. Different types of structure: (a) Penrose structure, concentration of small Robinson triangles, $c_p \approx 0.382$; (b) a twin structure ($c = 0.5$, five twins); (c) a random structure ($c \approx 0.37$); (d) a precipitated structure ($c = 0.8$). The fictitious atoms should be placed in the corners of the Robinson triangles (●). All possible connections between 'atoms' of lengths 1 and τ have been marked and one arbitrary choice of connected small triangles is hatched.

and Orland 1986, Stern *et al* 1986, Wolny *et al* 1988, Lebech *et al* 1988). As described by Wolny *et al* (1988), these structures can generally be divided into four types (figure 1): Penrose-like structures, twins, random structures and precipitated structures. All of them give tenfold diffraction patterns with well defined diffraction peaks. After averaging the peak intensities over the five equivalent directions and normalising to N^2 , where N is the number of 'atoms' in the structure, we find that for finite structures the peak intensities approach unity for large values of scattering vectors.

We limit the discussions to include only diffraction peaks for which the intensities when normalised to N^2 are greater than 0.1. We also use the term 'quasi-crystal' to describe structures having diffraction patterns exhibiting forbidden symmetry and having normalised peak intensities which are constant as a function of N . In other words it is only the symmetry that distinguishes the diffraction patterns of normal crystals and 'quasi-crystals'. It should be noted that this use of the term 'quasi-crystals' is unconventional because the term quasi-crystals is more conventionally used to describe all structures having well defined peaks in the diffraction patterns exhibiting forbidden symmetry, even those for which the normalised intensities are not necessarily constant with increasing number of atoms.

Figure 2 shows the diffraction patterns for the four structures mentioned above, computed along the k_y direction for $k_x = 0$. From this figure it can be seen that the series of peaks are completely aperiodic on a linear scale (k_y). However, all the diffraction peaks can be divided into groups of peaks such that each group becomes periodic on a logarithmic scale with the periodicity $\ln \tau$, where τ is the golden mean value equal to $(1 + \sqrt{5})/2$. The origin of such groups of peaks has been shown analytically by Jarić (1986). For each group of peaks the parallel and perpendicular components of the Q_m vector in the five-dimensional hypercubic reciprocal space are given by: $Q_m^{\parallel} = \tau^m Q_m^{\perp}$,

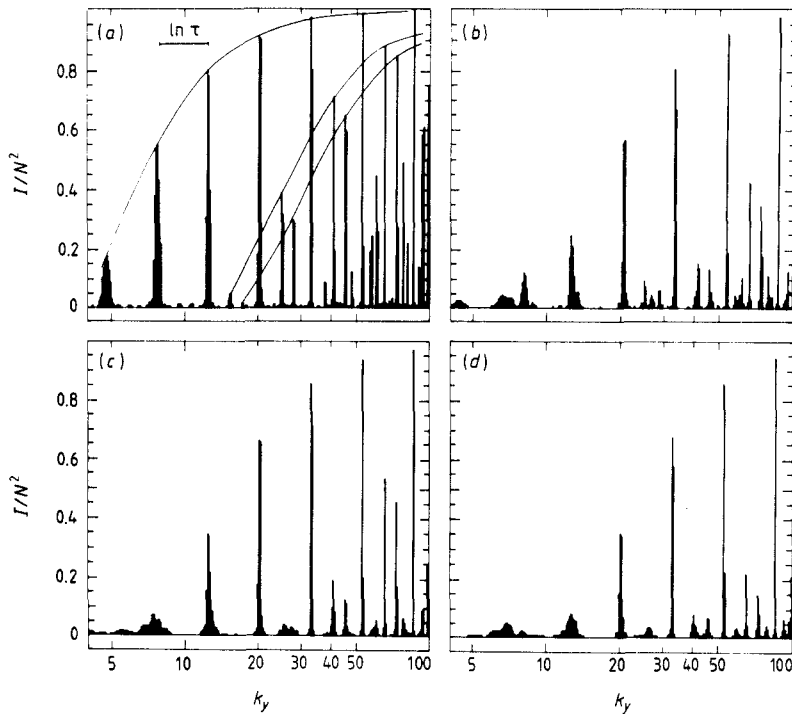


Figure 2. Normalised diffraction patterns in the k_y direction of reciprocal space for (a) Penrose tiling, (b) a twin structure, (c) a random structure and (d) a precipitated structure. On a logarithmic k_y scale all the peaks can be divided into series of peaks with a periodic arrangement of peaks. The period is equal to $\ln \tau$ within each series. In (a) the peaks in the first three series of peaks are joined by the thin curves.

$Q_m^- = (-\tau)^{-m} Q^\perp$ and $Q_m^{+z} = (-2)^m Q^{\perp z}$ for the m th peak in the group. Moreover, for all the structures discussed, a universal envelope function (or scaling function), appropriately shifted on a logarithmic k_y scale, describes the peak intensities for all the peaks in a particular series of peaks (figures 2(a) and 3). The shape of the envelope function can be computed as done by Jarić (1986), provided that the details of the hypersurface used in the projection method, namely its coordinate along the $[1, 1, 1, 1, 1]$ direction and its window function, are given. Computations using a Gaussian window function give a shape which is very close to our numerical results. The universal behaviour of the envelope function can be used to obtain parameters which may be regarded as similarity parameters. These parameters depend only on the type of structure and on the concentration c of small Robinson triangles in the structure and they describe the similarity of a given finite structure to the Penrose structure. As examples, the Penrose structure corresponds to $c = 1/\tau^2$, while $c = 0.5$ corresponds to a structure with equal numbers of small and large triangles.

2. Definition of the similarity parameters

Similarly to classical crystallography, the maximum peak intensities when normalised to N^2 do not depend on N for the Penrose-like structures, which are 'quasi-crystals'. This is in contrast with the behaviour of other types of structure for which the normalised peak intensities decrease to zero when N goes to infinity (non-'quasi-crystals'). Furthermore, the intensities vary for different groups of peaks (Wolny *et al* 1988) for the

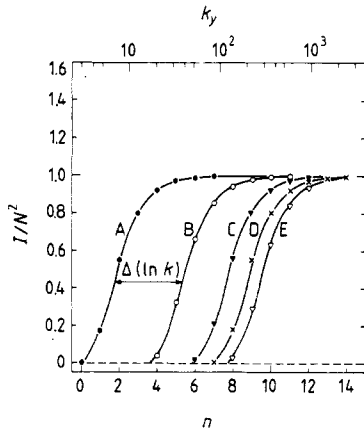


Figure 3. Peak intensity against the order n of the diffraction peak and/or $\ln k_y$ for one series of peaks described by $k_x = 0, k_y = k_0 \tau^n$, where $k_0 = 2\pi/d$ and $d = (2 + \tau) \sin(36^\circ)$, which gives $k_0 = 2.955$: curve A, Penrose tiling ($c = c_p = 1/\tau^2 \approx 0.382$); curve B, random structure ($c = 0.37$); curve C, twin structure ($c = 0.5$); curve D, precipitated structure ($c \approx 0.8$); curve E, twin structure ($c = 1.0$). A single envelope function appropriately shifted by $\Delta \ln k = \Delta n \ln \tau$ scales all peaks for the different structures. The same envelope function also scales peaks for other series of peaks described by a different value of k_0 .

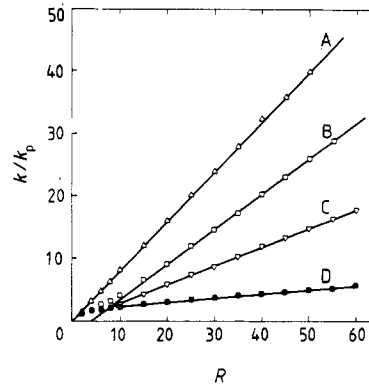


Figure 4. Scaling factor k/k_p for circular segments of a structure of radius R : curve A, twin structure ($c = 1.0, k_0 = 2.95$); curve B, precipitated structure ($c \approx 0.8, k_0 = 2.95$); curve C, twin structure ($c = 0.5, k_0 = 2.95$); curve D, random structure ($c = 0.37, k_0 = 2.95; k_0 \approx 9.56$ and $k_0 \approx 10.69$). This ratio was calculated as $k/k_p = \tau^{\Delta n}$, where Δn is the shift between the envelope function for a certain series of peaks in a given structure and the same series of peaks of Penrose tiling (see figure 3).

non-Penrose structures. However, as mentioned previously, for a given type of finite structure with a particular concentration of small triangles the wavevector dependence (in a logarithmic scale) of the intensities of each group of peaks is the same as for the Penrose structure apart from a shift ($\Delta \ln k = \Delta n \ln \tau$) along the horizontal axis (see figure 3). This characteristic property of the diffraction patterns leads to the main conclusion that for finite structures we may regard the Penrose tiling as the reference structure and relate all other structures discussed here to this particular structure. In other words, for all groups of peaks belonging to a given finite structure there is a universal scaling factor k/k_p , which causes the envelope of the intensities to coincide with the envelope obtained for the Penrose structure, although the peaks do not necessarily coincide. The scaling factor is strictly related to the value of the shift of Δn (or $\ln k$) by the following relation:

$$k/k_p = \tau^{\Delta n}$$

where Δn is the shift of the envelope function defined in figure 3. It should be noted that the above conclusion is not valid for rather weak peaks, i.e. peaks having maximum intensities less than 0.1, but, as mentioned previously, such peaks are excluded in the present analysis.

To study the spatial variation of the scaling factor we have computed k/k_p for two types of structure segment, i.e. for circles of variable radius R (figure 4) and for rings of mean radius R and thickness $\Delta R = 2$ (figure 5). For large values of R , both values of scaling factors are linear in R , with a ratio between the slopes of approximately $\frac{2}{3}$. This ratio is easily explained: let us assume the linear dependence for the ring-shaped structure

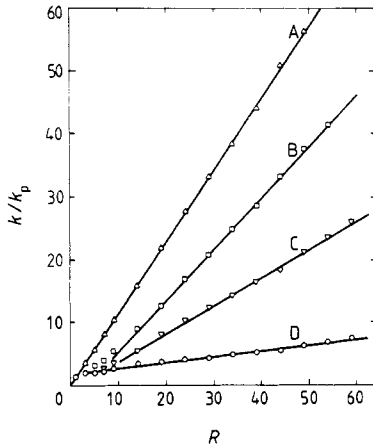


Figure 5. Scaling factor k/k_p for ring segments of structures of mean radius R and thickness $\Delta R = 2$ (this ratio was calculated as described for figure 4): line A, twin structure ($c = 1.0$, $k_0 \approx 2.95$); line B, precipitated structure ($c = 0.8$, $k_0 \approx 2.95$); line C, twin structure ($c = 0.5$, $k_0 \approx 2.95$); line D, random structure ($c = 0.37$, $k_0 \approx 2.95$, 9.56 and 10.69).

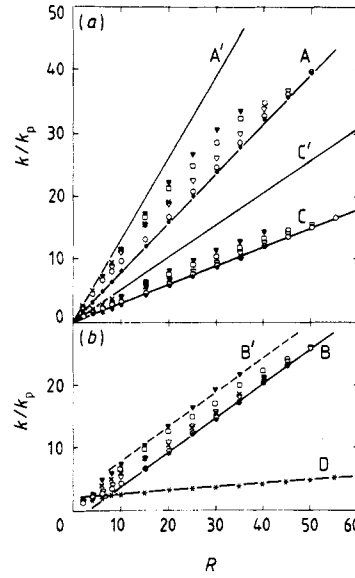


Figure 6. Scaling factor k/k_p for circles of radius R centred at different points (x, y) (\bullet , $(0, 0)$; \circ , $(-5, 0)$; ∇ , $(-10, 0)$; \square , $(-15, 0)$; \blacktriangledown , $(-20, 0)$; \times , $(0, 10)$) where $(x, y) = (0, 0)$ corresponds to the central points of the structures shown in figure 1: line A, twin ($c = 1.0$); line C, twin ($c = 0.5$); line A', single crystal grain ($c = 1.0$); line C', single crystal grain ($c = 0.5$); lines B, B', precipitated structures ($c = 0.8$); line D, random structures ($c = 0.37$), independent of (x, y) .

in figure 5 to be $k/k_p = A_R R + B_R$; then by simple integration we can find that the average value of k/k_p for the circle is also linear with R :

$$\frac{1}{\pi R^2} \int_0^R (A_R r + B_R) 2\pi r \, dr = \frac{2}{3} A_R R + B_R = A_c R + B_c$$

and the corresponding coefficients are given by

$$A_c = \frac{2}{3} A_R \quad B_c = B_R$$

in consistency with figures 4 and 5.

It turns out that, for a given type of structure defined by a particular set of matching rules (or concentration when the structure is random), its similarity to the Penrose structure can be described by the coefficients obtained from plots such as figures 4 or 5. The parameter A in this linear dependence describes the deviation from the classical scaling of the intensity. For ordinary crystals and 'quasi-crystals' the maximum peak intensities when normalised to N^2 are constants as a function of N , which corresponds to A equal to zero. Whenever A differs from zero, it means that there are some twins or microtwins in the structure with overlapping peaks (Wolny *et al* 1988). The parameter A describes the space dependence of Penrose-like order and it can be used to measure the deviation from a 'quasi-crystal' structure. The parameter B can be used to distinguish between 'quasi-crystals' with different degrees of Penrose-like order, as discussed later. In the analytical works on the equilibrium random tilings the fluctuation of the perpendicular coordinates of the five-dimensional hypersurface is mostly used (Henley

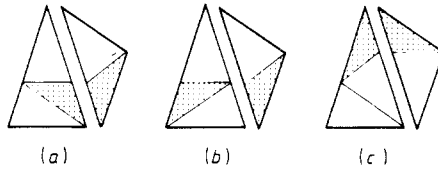


Figure 7. Robinson triangles and the divisions that may be used in the inflation method.

1988). It is in some way equivalent to our scaling parameter A , because it controls the intensities of the peaks in the diffraction pattern. For such a variable the dependence on R should have the form of a logarithmic function, provided that there is no uniform phason strain in the structure (Tang and Jarić 1989). In our structures obtained by a growth model the logarithmic component is not visible. The possible explanations are that linear dependence is dictated by the uniform phason strain present in our samples and the A parameter is closely related to the strain value or that the structures differ from equilibrium random tilings.

Figure 6 shows plots similar to those shown in figure 4 for different positions of the origin of the circle considered in the calculation. It is obvious that the random structure is the least sensitive to a change in the origin (figure 6(b)). Precipitated structures are heterogeneous and the local concentration of triangles changes considerably with change in the origin. This results in a scatter of the points in the curve describing the behaviour of the precipitated structures (figure 6(b)). In figure 6(a) we show the results obtained for two twin structures. In one case, the concentration of small triangles is 0.5, which corresponds to the structure shown in figure 1(b). In the other case, the concentration is unity because the structure was generated using only small triangles. In figure 6(a) the lines A and C describe the behaviour for the origin of the circle in the centre of the five fold symmetry (see figure 1(b)) for concentrations of 0.5 and 1, respectively. Lines marked A' and C' describe the behaviour for the origin of the circle shifted far in the x direction from the centre of the fivefold symmetry into the interior of one of the grains. When symmetrised over the five directions, the fictitious 'diffraction patterns' of such a single grain also show groups of peaks similar to those found in the Penrose tiling. The non-linearity observed in figure 5(a) for the curves corresponding to shifted circles is connected with the influence of the inter grain boundaries (a change in the matching rules).

3. Disorder and the similarity parameters

To find the influence of disorder on the similarity parameters, we calculated k/k_p for two different classes of random structures. To the first class belong all the structures which can be generated using the inflation method (Wolny *et al* 1988). As seen from figure 7, one can easily subdivide the triangles in several different ways, but for each choice of subdivision the concentrations are the same and finally go to $c_p = 1/\tau^2$. The subdivision shown in figure 7(a) leads to the Penrose tiling shown in figure 1(a). It is impossible to tile the plane using only the subdivision shown in figure 7(c); to avoid defects, one has to use it together with the other ways of subdividing. If the subdivision of figure 7(c) is preferred, one obtains the structure shown in figure 8(a). If the triangles are subdivided by a random choice between the different ways shown in figure 7, there are many random structures which tile the plane without defects, with concentration of

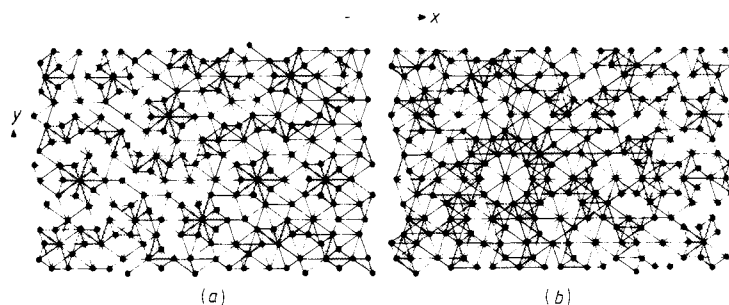


Figure 8. Different types of structure obtained by the inflation method: (a) the division shown in figure 7(c) was preferred; (b) the different divisions shown in figure 7 were used at random.

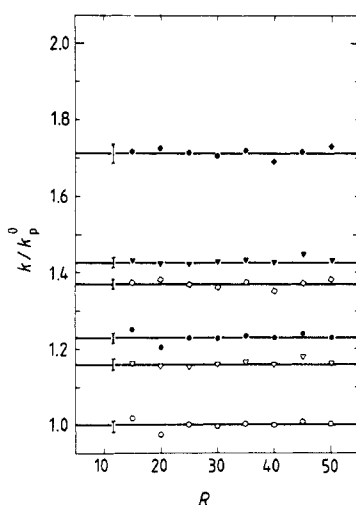


Figure 9. Scaling factor k/k_p^0 for ring segments of structures of mean radius R and thickness $\Delta R = 2$ for the first two series of peaks described by the relation $k = k_0 \tau^n$ (k_p^0 is the apparent scattering vector for the first series of peaks ($k_0 \approx 2.955$) of the Penrose tiling; if instead k_p was used as it is in figure 5, the lines for the different series would coincide for $k_0 \approx 2.95$ (\circ, ∇, \diamond) and for $k_0 \approx 9.56$ ($\bullet, \blacktriangledown, \blacklozenge$): \circ, \bullet , Penrose structure (figure 1(a)); $\nabla, \blacktriangledown$, random Penrose structure (figure 8(b)); \diamond, \blacklozenge , structure shown in figure 8(a) obtained by using the preferred division of triangles shown in figure 7(c).

small triangles approaching c_p . One of these structures is shown in figure 8(b). The calculated diffraction patterns for all these structures look similar to that obtained for Penrose tiling and the similarity parameters A and B can be obtained from figure 9. In this figure we relate two series of peaks (for $k_0 \approx 2.95$ and $k_0 \approx 9.56$) to the first series ($k_0 \approx 2.95$) of the Penrose tiling. It is evident from figure 9 that, for these series (and, of course, also for others) and for all structures obtained by the inflation method, the parameter A (slope) is equal to zero whereas the parameter B is different for the different structures. The fact that the intensities are independent of the sample dimensions R means that all these structures belong to the same type of structures, called 'quasi-crystals' in this paper. The value of B for one series of peaks ($k_0 \approx 2.95$) is the same as for the other series of peaks ($k_0 \approx 9.56$) and the different relative values of B describe different levels of disorder in these structures. It should be noted, however, that for the first peaks in each series (which correspond to high Q^\perp values) a weak dependence of peak intensities on the number (N) of 'atoms' can be seen although it does not influence the values of A significantly.

The second class of random structures is represented by the random structure shown in figure 10(b). This structure was obtained by random mixing of the two different twins shown in figures 1(b) and 10(a). Each of them has $c = 0.5$ but they are generated using

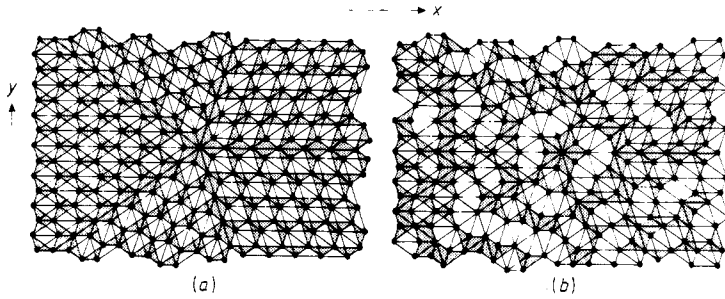


Figure 10. (a) Twin structure ($c = 0.5$); (b) random twin structure ($c = 0.5$) obtained by random mixing of the local configurations used in the structures shown in figures 1(b) and 10(a).

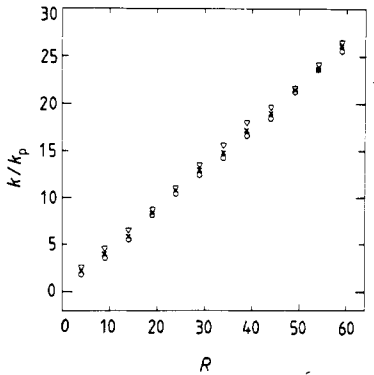


Figure 11. Scaling factor k/k_p for different structures with the concentration c of small Robinson triangles equal to 0.5: \circ , twin structure from figure 1(b), $A = 0.441 \pm 0.007$; ∇ , twin structure from figure 10(a), $A = 0.440 \pm 0.006$; \times , random twin structure from figure 10(b), $A = 0.443 \pm 0.004$.

different matching rules. From figure 11 it is quite evident that the randomisation does not influence the slope of the similarity parameter A . From the two examples discussed above, the general conclusion can be reached that the similarity parameter A depends only on the concentration of small triangles. The random arrangement of triangles has no influence on this parameter.

4. Concentration and the similarity parameter A

We have investigated several structures built using different concentrations of small Robinson triangles and determined the concentration dependence of the similarity parameter A . We mainly studied this dependence for twins, some of which are shown in figures 1(b), 10(b) and 12, but also for the structures described by Wolny *et al* (1988), i.e. random structures (see also figure 1(c)) and precipitated structures (figure 1(d)). The dependence of the parameter A on the concentration is shown in figure 13. It shows a singular point for the concentration of small triangles equal to c_p , i.e. the Penrose tiling concentration. All the structures for this particular concentration belong to crystals which we call the 'quasi-crystals', i.e. structures with diffraction patterns exhibiting forbidden symmetry, but with the same scaling of the peak intensities as in classical crystallography. For these structures the similarity parameter A equals zero. Structures having concentrations that differ from c_p lie on the common curve (figure 13) irrespective of their type (perfect twins, random or precipitated structures). This means that all those structures belong to the same class of structures which may be called twins or microtwins.

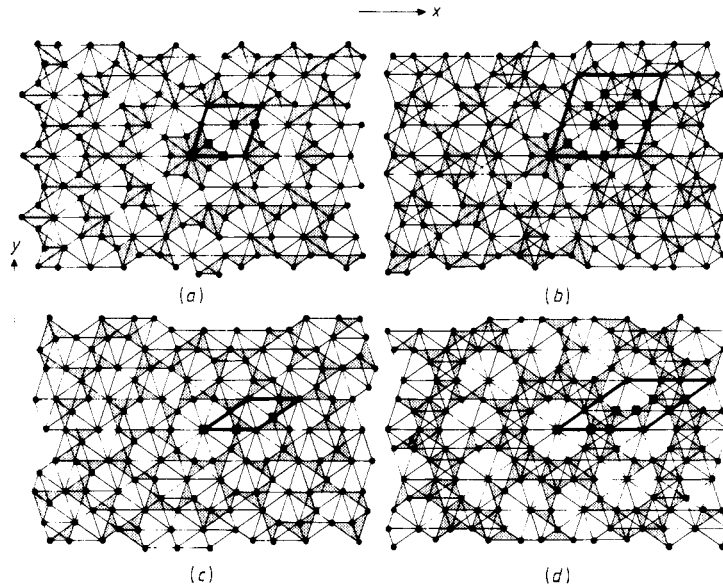


Figure 12. Twin structures for different concentrations of small Robinson triangles: (a) five twins, five 'atoms' per unit cell, four small and six large Robinson triangles per unit cell, $c = 0.4$; (b) five twins, 13 'atoms' per unit cell, 10 small and 16 large Robinson triangles per unit cell, $c = 0.3846$; (c) 10 twins, three 'atoms' per unit cell, two small and four large Robinson triangles per unit cell, $c = 0.3333$; (d) 10 twins, eight 'atoms' per unit cell, six small and 10 large Robinson triangles per unit cell, $c = 0.375$. The 'atoms' in one unit cell are indicated (■).

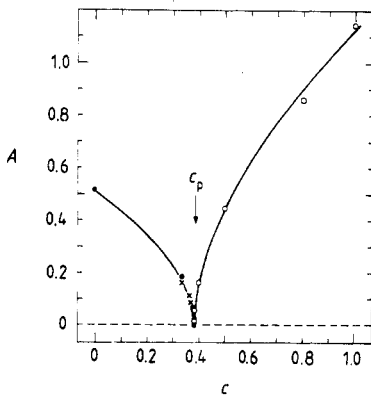


Figure 13. Dependence of the similarity parameter A on the concentration c of small Robinson triangles for different types of ring shape structure: ○, ●, different twin structures; ×, random structures with different concentrations of small Robinson triangles; □, precipitated structure ($c = 0.8$); ■, Penrose structure; —, two curves given by the formula $A = A_0(\Delta c)^{\alpha}$, where $\Delta c = c - c_p$, $c_p = 1/\tau^2$ and the critical exponent is equal to (0.55 ± 0.02) . The deviation of the precipitated structure from the full curve can be explained by the concentration fluctuation observed for this structure.

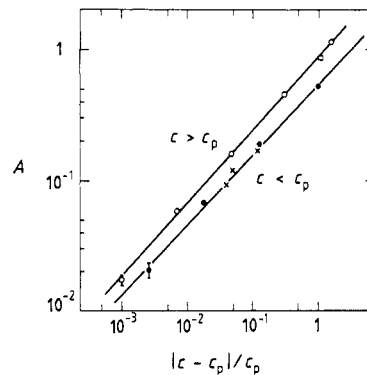


Figure 14. Log-log plot of the similarity parameter A against the reduced concentration of small triangles: ○, ●, twins; □, precipitated structure; ×, random structure. Typical critical behaviour is observed with critical exponents equal to 0.539 ± 0.018 (for $c < c_p$) and 0.558 ± 0.007 (for $c > c_p$) and the ratio between the pre-exponential factors equal to $1.6 (\approx \tau)$.

The random structures of Wolny *et al* (1988) are examples of microtwins; the existence of very small twins in the random structure can also be seen in figure 1(c). For these microtwins the complex peaks observed at Penrose positions cannot be split into individual peaks by increasing N , a behaviour which is observed for the normal twins (Wolny *et al* 1988). The singular behaviour of the similarity parameter A indicates a kind of continuous transition from twins (or microtwins) to 'quasi-crystals' at the critical concentration c_p . From figure 14 a critical exponent describing the behaviour of the similarity parameter A is found to be 0.55 ± 0.02 .

5. Conclusions

There are two types of structure which tile the plane without defects and for which the diffraction patterns can show symmetry elements forbidden by classical crystallography, such as fivefold or tenfold symmetry. To one of these types belong all structures for which the peak intensities are constant when normalised to N^2 , as it is for normal crystals. We call these structures 'quasi-crystals'. The structures for which the peak intensities do not scale as N^2 belong to the second class of structures, and these structures may be called twins or microtwins. The microtwins differ from the twins by showing a different behaviour with complex peaks at Penrose positions. For twins these peaks can always be split into individual peaks by increasing N ; this is never observed for microtwins (Wolny *et al* 1988). Analytical results for equilibrium random tilings (Strandburg *et al* 1989, Widom *et al* 1989) show that the intensities of the peaks should scale as $N^{2-\eta/2}$ where $\eta = |Q^+|^2/2\pi k + |Q_z^+|^2/2\pi k_z$. Here k and k_z denote the phason stiffness constants and Q^+ and $|Q_z^+|$ the perpendicular components of Q in the five-dimensional hypercubic reciprocal lattice for a given peak. For our random structures obtained by the growth model the peak intensities do not scale with N according to that formula unless it is assumed that the η -value changes with the size of the sample.

For all finite structures which tile the plane without defects using only two types of decorating element, the diffraction patterns exhibit the same series of peaks as those observed for the corresponding Penrose tiling. Using Penrose tiling as a reference structure and the universal behaviour of a function describing the change in peak intensities against $\ln k$ —the envelope function—one can easily find a linear relationship between the scaling factor k/k_p and the dimension of a circular segment of the sample structure. From plots such as those presented in figure 4 or 5 for a given structure, the two parameters (A and B) of the linear dependence of the scaling factor can be obtained. The parameter A distinguishes 'quasi-crystals' ($A = 0$) from twins or microtwins ($A \neq 0$). This parameter is the same for the ordered and disordered (random) structures having the same concentration of small triangles.

For all investigated structures the parameter A shows a common dependence on the concentration (figure 13) of small Robinson triangles, having a singular point at $c = c_p$. This behaviour indicates a kind of continuous transition from one type of structure (twins or microtwins) to the other ('quasi-crystals'). The critical exponent for the parameter A for this transition is equal to 0.55 ± 0.02 .

It has been shown by Henley (1988) and Tang and Jarić (1989) that for the equilibrium random tilings the deviation from the critical concentration is related to the uniform phason strain and that this dependence does not include the linear term. In such a case the uniform phason strain value should go as $(c - c_p)^{0.5}$ provided that the quadratic term dominates the dependence. The value of the critical exponent obtained for our structures strongly supports the conjecture that the A parameter is proportional to the phason

strain. The deviation from 0.5 can be explained by the influence of higher-order terms in the relation between the concentration deviation and the phason strain or by the non-equilibrium nature of our structures.

For real structures the way to classify the structure as one of the two types ('quasi-crystals'; twins or microtwins) is to make a plot like either figure 4 or figure 5. If the peak intensities scale as N^2 , the structure is a 'quasi-crystal'; if they do not scale as N^2 , the structure is a twin or a microtwin. Because the 'quasi-crystals' are limited only to a certain concentration (at least in two dimensions), we rather expect that most real structures would be twins or microtwins. However, experimental verification could be difficult, especially for concentrations close to the critical point.

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