Mistakes in Quasilattices

Jian Ping Lu and Joseph L. Birman
Department of Physics, City College of the City University of New York, New York, New York 10031
(Received 18 February 1986)

We studied a class of mistakes or faults in quasilattices. The effect of a random distribution of mistakes on the diffraction of 1D, and a special class of 3D, quasilattices is calculated exactly. Mistakes change the diffraction pattern qualitatively: Some Bragg peaks decrease in intensity as expected, but some are enhanced. As a result some spots disappear and some new ones appear. The diffuse scattering is also calculated. Results are given comparing calculated diffraction patterns in fivefold, threefold, and twofold symmetry directions for a 3D quasicrystal with and without mistakes.

PACS numbers: 63.50.+x, 05.50.+q

Perfect quasilattices.—Several workers have calculated the x-ray diffraction pattern of perfect quasicrystals,[1] with use of the Penrose tiling as the basic model structure, and have obtained good qualitative agreement with experiments.[2] There are also various studies of perfect Penrose lattices in different dimensions.[3-6] In this Letter we discuss a special class of defect, namely *mistakes* in a quasilattice (QL). We study the effect of mistakes on the diffraction intensity in a one-dimensional quasiperiodic lattice exactly; then we generalize to higher dimension. Significant qualitative changes occur in diffraction intensities when mistakes are present. Real quasicrystals are obtained by highly nonequilibrium processes, imperfection is inevitable, and mistakes have now apparently been observed.[7]

Consider a perfect one-dimensional QL, namely the Fibonacci lattice (FL),[2,6] defined as a 1D tiling with two basic unit lengths $S=1$ and $L=\tau=(1+\sqrt{5})/2$. The position of the nth lattice point $x(n)$ is

$$x(n)=n+\alpha+[n/\tau+\beta]/\rho; \tag{1}$$

$0\le\alpha,\beta<1$, and $\rho>1$ are three parameters. Here we take $\rho=\tau$ for simplicity.[6]

A difference between quasiperiodic, periodic, and random lattice is the configuration number for n sites, $C(n)$, which is the number of distinct configurations for an arbitrary n segment. $C(n)$ is constant for the periodic lattice, and exponential in n for a random lattice. For the QL, $C(n)$ is linear in n. For FL, $C(n)=n+1$: There are only three two-segment and four three-segment configurations. The segments SS and LLL are prohibited (Fig. 1).

Mistakes in a one-dimensional quasilattice.—For the FL, one of the simplest types of defect is transposition in sequential order of L,S segments. If we impose a restriction that the random occurrence of transpositions will not completely violate the geometrical properties described above, the energy cost is small.[8] One expects that such a type of defect is generic. Since it differs from the usual defect discussed in crystals, and is similar to mistakes in stacking sequence discussed by Wilson and others,[9] we call it a *"mistake."*

The simplest restriction requires that the three-site configuration remain unchanged, i.e., maiantain the following selection rules: no SS and no LLL. Careful examination of the FL shows that possible mistakes consistent with these constraints have the form of a transposition in the five-site configurations, $LLSLS\rightarrow LSLLS$ (Fig. 1); one has to exclude $SLSLL\rightarrow SLLSL$.

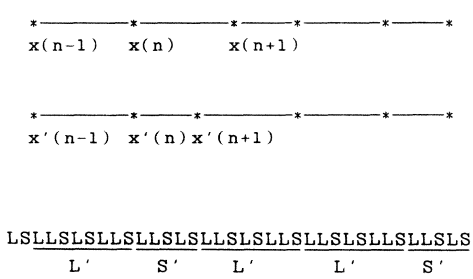
Label the position of the nth lattice site in the imperfect FL as $x'(n)$. Suppose a mistake occurred at site n, namely

$$\begin{aligned} &x'(n-1)=x(n-1);\\ &x'(n)=x'(n-1)+S;\\ &x(n)=x(n-1)+L;\\ &x'(n+1)=x'(n)+L=x(n+1);\\ &x(n+1)+x(n-1)+L+S. \end{aligned} \tag{2}$$

This changes the Fourier transform from that of the perfect lattice $F(q)$ to that of an imperfect lattice denoted $F'(q)$: The term $\exp[iqx(n)]$ is replaced by

$$\exp[iqx'(n)]=\exp[iqx(n)]-\exp[iqx(n-1)]\Delta,$$

where $\Delta=\exp(iqL)-\exp(iqS)$.



(a) (b) (c)

FIG. 1. (a) Cluster $LLSLS$ in the perfect FL. (b) A mistake occurred at site $x(n)$: The cluster became $LSLLS$. (c) A segment of a perfect FL. All clusters of type $LLSLS$ are underlined. The distance between these clusters is either $S'=3L+2S=\tau^4S$ or $L'=5L+3S=\tau^4L$ and follows a Fibonacci sequence; hence form a FL with basic length $S'=\tau^4S$.

For every *LLSLS* cluster in the perfect FL, impose a probability p that it will flip into *LSLLS*. Label the position of these clusters by $y(m)=x(m-1)$, where $x(m-1)$ is the position of the atom which is between the first two long spacings of the cluster (Fig. 1). Note that the set $y(m)$ is only a subset of all $x(n)$. The Fourier transform of the lattice with a given configuration of mistakes is

$$F'(q,\{\sigma\})=\sum_{n=-\infty}^{\infty}\exp[iqx'(n)]=\sum_{n}\exp[iqx(n)]-\sum_{y(m)}\exp[iqy(m)]\Delta(\sigma_m), \tag{3}$$

where

$$\Delta(\sigma_m)=\exp(iqS\sigma_m)-\exp(iqS);\quad \sigma_m=\begin{cases}\tau=L/S & \text{with probability } p,\\ 1 & \text{with probability } (1-p).\end{cases} \tag{4}$$

Assume that mistakes occur independently with probability p at all possible sites $y(m)$. To obtain the observed diffraction intensity $I(q)=\langle FF^*\rangle$ one has to average over all possible configurations of $\{\sigma\}$. In general,

$$\langle F(q,\{\sigma\})F^*(q,\{\sigma\})\rangle=\langle F(q,\{\sigma\})\rangle\langle F^*(q,\{\sigma\})\rangle+D(q); \tag{5}$$

$D(q)$ is the diffuse scattering. To calculate the first term, define

$$F'(q)\equiv\langle F'(q,\{\sigma\})\rangle=F(q)-\langle f(\{\sigma\})\rangle. \tag{6}$$

Here $f(\{\sigma\})$ is the second term in Eq. (3) and is a function of configuration $\{\sigma\}$:

$$\langle f(\{\sigma\})\rangle=\sum_{y(m)}\exp[iqy(m)]\langle\Delta(\sigma_m)\rangle=p\Delta\sum_{y(m)}\exp[iqy(m)], \tag{7}$$

with $\Delta=\Delta(\tau)$. Careful analysis of all possible sites reveals that the set $y(m)$ itself forms a new FL with basic lattice length $S'=3L+2S=\tau^4S$, and a possible shift of origin (Fig. 1). Therefore

$$F'(q)=F(q)-p\Delta F(\tau^4q)\exp(iq\eta), \tag{8}$$

where η is a constant depending on (α,β) and the exact procedure one used to define the scaling. But the phase is not relevant to our results on the effect of mistakes on $I(q)$.[8]

The diffraction function of the perfect FL has been calculated by several workers.[2,8] The known result is that $F(q)$ consists of a sum of weighted δ functions dense in q space:

$$F(q)=\sum_{m,n}\exp(i\psi_{mn})[\sin z_{mn}/z_{mn}]\delta(qa-q_{mn}a), \tag{9}$$

where $q_{mn}=2(\pi/\sqrt{5})(n\tau+m)$; $z_{mn}=2(\pi/\sqrt{5})(n-m\tau)$; $\psi_{mn}=q_{mn}\alpha+(2\beta-1)z_{mn}$; and $a=\sqrt{5}/\tau$ is the average lattice spacing (for simplicity we have taken $S=1$). Substituting Eqs. (9) and (10) into Eq. (8), after some algebra one has

$$F'(q)=\sum_{m,n}\frac{\exp(i\psi_{mn})\sin(z_{mn}/2)-p\sin(q_{mn}/2\tau)\exp(i\hat{\psi}_{mn})\sin(z_{mn}/2\tau^4)}{z_{mn}/2}\delta(qa-q_{mn}a), \tag{10}$$

where

$$\hat{\psi}_{mn}=q_{mn}\tau^4\alpha+(2\beta-1)\tau^{-4}z_{mn}+q_{mn}/2\tau+\pi/2.$$

The second term in Eq. (5) has been calculated exactly in Ref. 8:

$$D(q)=[4p(1-p)\tau^{-4}/N]\sin^2(q/2\tau), \tag{11}$$

where N is the linear dimension of the crystal in units of the average atomic spacing a. Experimentally, a typical diffraction peak line width Δq is about 10^{-2} in units of $1/a$, and so the contribution of $D(q)$ to the peak intensity is order of $(p\,\Delta q)/(N\tau^4)$. Presently available samples have $N=10^3$ in a single grain; so for $p=0.2$ (Fig. 2) the contribution is order of 10^{-6}. Therefore for all diffraction peaks whose intensity is larger than 10^{-6} of the maximum peak intensity, the diffuse background can be neglected. On the other hand, those peaks whose intensity ratio to the maximum is of order 10^{-6} or smaller will merge into smooth background as a result of diffuse scattering. This will be hard to distinguish from other incoherent scattering. In this Letter we are primarily interested in those peaks whose intensity is around a cutoff which is taken to be much larger than the contribution from the diffuse term, and so $D(q)$ will be neglected in the following calculations.

From the above equations one sees that mistakes change the relative intensities of different Bragg peaks, depending on the values of p and q_{mn}. As expected, some peaks decrease in intensity. An unexpected result is that some peaks are enhanced. This effect is more pronounced in the three-dimensional case, as we show below.

The restriction we have imposed on the imperfect QL

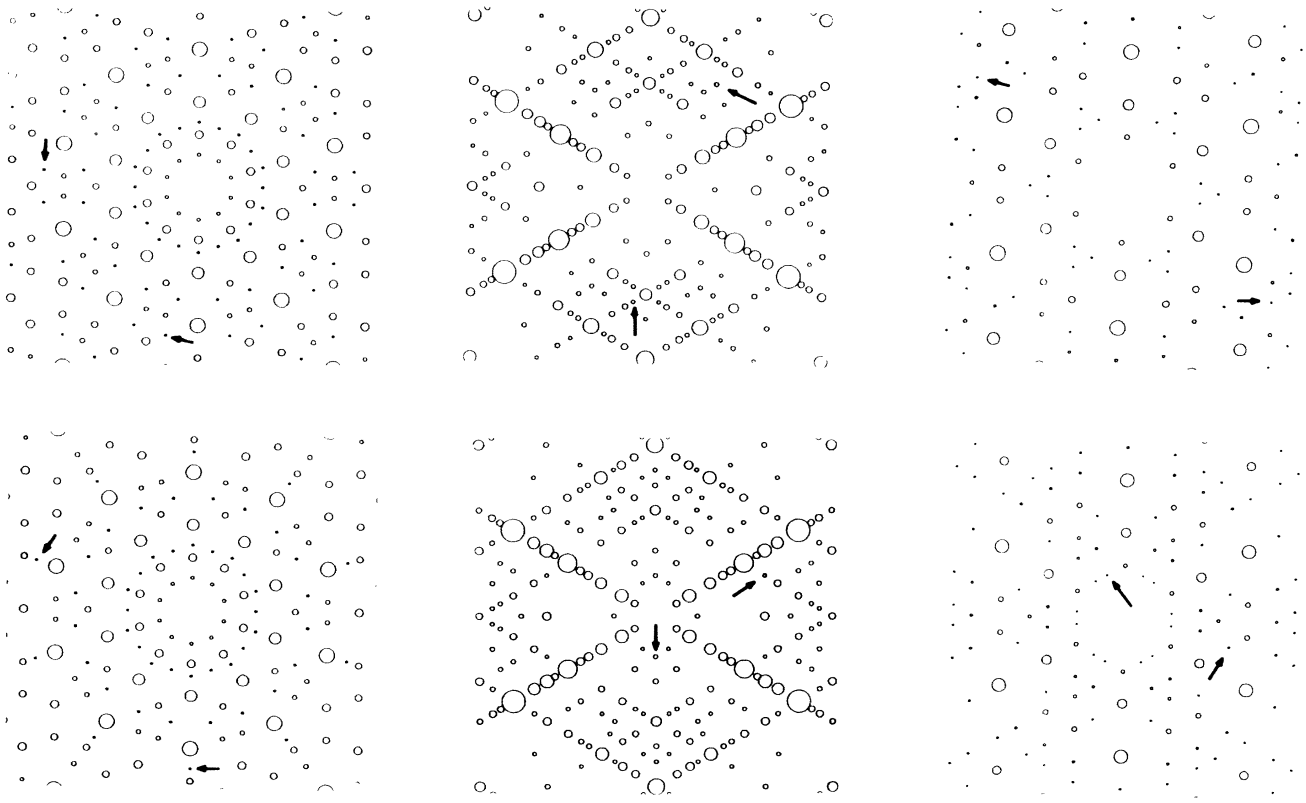


FIG. 2. The diffraction pattern calculated from Eqs. (8) and (10) with $p=0.2$ (the "concentration" of mistakes is $p\tau^{-4}=3\%$) in Eq. (11). The radius of a spot corresponds to the intensity. Top row: perfect lattice; bottom row: imperfect case. When mistakes occur, spots such as those shown by arrows in the top row are diminished below cutoff, and spots such as those shown by arrows in the figures in the bottom row are enhanced above the cutoff. Shown is diffraction on planes normal to (a) fivefold, (b) twofold, and (c) threefold axes.

could be weakened, e.g., by our allowing more general types of permutation, namely by permitting the variable σ_m in Eq. (4) to have a continuous distribution. This will not change the overall calculation; however, now the Δ will have a more complicated dependence on q. Another way of generalizing is to require that the n-site configuration be kept the same. Then instead of τ^4 in Eq. (11) a different power of τ may appear. For example if one permits every LS in the Fibonacci sequence to transpose to SL, one will get τ^2 scaling.

Mistakes in a three-dimensional quasilattice. —Qualitatively one expects that the same effects will occur in a three-dimensional quasiperiodic structure, namely in the generalized Penrose tiling. Levine and Steinhardt[3] have shown that for the Ammann QL, the Fourier transform is a simple product of the Fourier transforms of three 1D FL. Since the skeleton of a three-dimensional Penrose tiling is equivalent to an Ammann QL, the qualitative result is also true for a general QL. Mathematically an Ammann lattice is a quasiperiodic hexagrid which is the simple direct product of six one-dimensional quasiperiodic grids, each with grid direction along one of six fivefold symmetry axes ($\mathbf{e}_i, i=0,1,\ldots,5$) of an icosahedron. Because the hexagrid is nonsingular (no more than three grids intersect at one point[3,4]), the diffraction amplitude $F_3(\mathbf{k})$ of the vertices of the hexagrid can be written as

$$F_3(\mathbf{q})=\sum_{i>j>k} F(\mathbf{q}\cdot\mathbf{u}_{ijk})F(\mathbf{q}\cdot\mathbf{u}_{jki})F(\mathbf{q}\cdot\mathbf{u}_{kij}), \tag{12}$$

where $F(x)$ is given by Eq. (9) and $\mathbf{u}_{ijk}=\mathbf{e}_j\times\mathbf{e}_k/[\mathbf{e}_i\cdot(\mathbf{e}_j\times\mathbf{e}_k)]$. Since the support of $F(q)$ is the set $q=q_{mn}$, the argument of each factor in Eq. (12) must be of the form q_{mn} in order that $F_3(\mathbf{q})$ not vanish. This requires $\mathbf{q}$ to be of the form

$$\mathbf{q}=(2\pi/a)[(n_i+m_i\tau^{-1})\mathbf{e}_i+(n_j+m_j\tau^{-1})\mathbf{e}_j+(n_k+m_k\tau^{-1})\mathbf{e}_k]. \tag{13}$$

We now extend the class of mistakes to the three-dimensional tiled lattice. Mistakes in tiling now correspond to a transposition of grid order. This kind of tiling is generated from the hexagrid by taking each grid sequence to follow the 1D imperfect FL. Therefore the diffraction amplitude $F_3'(\mathbf{q})$ of a QL with mistakes will be expressed in terms of $F'(q)$ of Eq. (10) as

$$F_3'(\mathbf{q}) = \sum_{i>j>k} F'(\mathbf{q}\cdot\mathbf{u}_{ijk})F'(\mathbf{q}\cdot\mathbf{u}_{jki})F'(\mathbf{q}\cdot\mathbf{u}_{kij}). \quad (14)$$

This diffraction intensity is qualitatively different from that of Eq. (12). As in the 1D case, some quasi-Bragg peaks decrease in intensity as expected, but some peaks are enhanced. In Fig. 2 we have plotted the results computed from Eq. (12) ($p=0$) and Eq. (14), taking $p=0.2$ in Eq. (11). Only those Bragg peaks whose intensity is larger than some arbitrarily chosen cutoff are shown. Results are that some Bragg peaks in the diffraction pattern of the perfect QL are extinguished, and that some Bragg peaks previously absent now appear above threshold. A few examples of each are shown with arrows in Fig. 2. This picture differs qualitatively from the diffraction pattern of a usual crystal with faults, where the effect of randomness always reduces the intensity of Bragg peaks.[9] These results are independent of the value chosen for the cutoff.

Figure 2 illustrates the symmetrical case, in which the shift parameter η [in Eq. (7)] is the same in all the grid directions. However, this is not necessary; mistakes can be implemented so that fivefold, twofold, and threefold rotational symmetry is lost, by our choosing different parameters for different grid directions. Results will be published elsewhere.

We thank Dr. C. L. Wang for help in the use of the Unix operating system, and Professor S. C. Moss for a preprint of Ref. 7. The work is supported by the Professional Staff Congress Grant No. PSC-FRAP-6-65280 and National Science Foundation Grant No. NSF-DMR-83-03981.

Note added.—After our work was completed we learned of important, and independent, work by Robertson, Misenheimer, Moss, and Bendersky[7] which includes a suggestion that faults occur in quasicrystals of Al-Mn-Si alloys. Their proposed faults are identical to the mistakes proposed and analyzed here.

[1]D. S. Shechtman, I. Blech, D. Gratias, and J. W. Cahn, Phys. Rev. Lett. **53**, 1951 (1984).

[2]D. Levine and P. Steinhardt, Phys. Rev. Lett. **53**, 2477 (1984); M. Duneau and A. Katz, Phys. Rev. Lett. **54**, 2688 (1985); P. A. Kalugin, A. Kitaev, and L. Levitov, Pisma Zh. Eksp. Teor. Fiz. **41**, 119 (1985) [JETP Lett. **41**, 145 (1985)].

[3]D. Levine, and P. Steinhardt, Phys. Rev. B **34**, 596 (1986).

[4]N. de Bruijn, Ned. Akad. Weten. Proc. Ser. A **43**, 39 (1981); P. Kramer and R. Neri, Acta Crystallogr. Sect. A **40**, 580 (1984).

[5]P. Bak, Phys. Rev. Lett. **54**, 1517 (1985); N. D. Mermin and S. M. Troian, Phys. Rev. Lett. **54**, 1524 (1985).

[6]J. P. Lu, T. Odagaki, and J. L. Birman, Phys. Rev. B **33**, 4809 (1986); R. Merlin, K. Bajema, R. Clarke, F. Y. Juang, and P. K. Bhattacharya, Phys. Rev. Lett. **55**, 1768 (1985).

[7]J. L. Robertson, M. E. Misenheimer, S. C. Moss, and L. Bendersky, Acta Metall. (to be published). See also K. Hiraga, M. Hirabayashi, A. Inoue, and T. Masumoto, J. Phys. Soc. Jpn. **54**, 4077 (1985).

[8]Jian Ping Lu and Joseph L. Birman, J. Phys. (Paris), Colloq., Supp. 7, C3-251 (1986).

[9]A. J. C. Wilson, *X-Ray Optics* (Methuen, London, 1962).