Strain Accumulation in Quasicrystalline Solids

Franco Nori, Marco Ronchetti,[a] and Veit Elser[b]
Institute for Theoretical Physics, University of California, Santa Barbara, California 93106
(Received 24 March 1988)

We study the relaxation of 2D quasicrystalline *elastic networks* when their constituent bonds are perturbed homogeneously. Whereas ideal, quasiperiodic networks are stable against such perturbations, we find significant accumulations of strain in a class of disordered networks generated by a growth process. The grown networks are characterized by root mean square phason fluctuations which grow linearly with system size. The strain accumulation we observe in these networks also grows linearly with system size. Finally, we find a dependence of strain accumulation on cooling rate.

PACS numbers: 62.30.+d, 61.70.−r

There is a widely held belief that in certain solids (e.g., glasses) there are large accumulations of strain even though external stresses are absent.[1] In crystalline solids the strain fields of isolated defects are well understood but fail to show any accumulation. In amorphous solids, where the phenomena is believed to exist, identification of an appropriate strain-free reference structure has always been a major problem.[2] Quasicrystalline solids[3] represent an interesting intermediate case in that the number of local structural elements is finite and yet these compose a structure with (possibly) positive configurational entropy. In this Letter, we exhibit a two-dimensional (2D) quasicrystalline model, a "decagon aggregate," where the phenomenon of strain accumulation is well defined. Numerical studies of our model show that the accumulation of strain is correlated with the behavior of the phason field.

The building blocks of our model are decagons packed edge to edge (see Fig. 1). In any aggregate of edge-sharing decagons, any two decagons can always be related by a pure translation. Moreover, such a translation can always be expressed as an integral linear combination of four basis vectors $\mathbf{e}_i^{\parallel}$ $(i=1,\ldots,4)$. The latter fact is equivalent to the statement that the possible decagon centers may be obtained by projecting a suitable 4D lattice. Consequently, each decagon is associated with a pair of two-component vectors: $\mathbf{x}^{\parallel}$, the location of its center; and $\mathbf{x}^{\perp}$, its "phason" coordinates. The pair $(\mathbf{x}^{\parallel},\mathbf{x}^{\perp})$ comprise the 4D lattice point. Details of the projection technique are given at length elsewhere.[4] For our purposes, it is sufficient to note that if the separation (in the physical plane) of two decagons is given by $\Delta\mathbf{x}^{\parallel}=\sum_{i=1}^{4} n_i\mathbf{e}_i^{\parallel}$, then $\Delta\mathbf{x}^{\perp}=\sum_{i=1}^{4} n_i\mathbf{e}_i^{\perp}$ gives the separation of their phason coordinates. The geometry of the $\mathbf{e}_i^{\perp}$ vectors is shown in the inset of Fig. 1.

The geometrical structure of edge-sharing decagons is our strain-free reference solid. We constrain the local structure by the requirement that the next-nearest-neighbor (nnn) decagon separation is $\tau=(1+\sqrt{5})/2$ times the nearest-neighbor (nn) or edge-sharing separation (see Fig. 1). If we depict only the pattern of nn bonds in the decagon aggregate, a network such as shown in Fig. 2 results. The connectivity of the network is significantly increased by including nnn bonds. Our model uses both types of bonds to stabilize the network mechanically. Specifically, we impose Lennard-Jones potentials between pairs of decagons joined by a bond. The Lennard-Jones scale parameter is given by σ_1 for nn

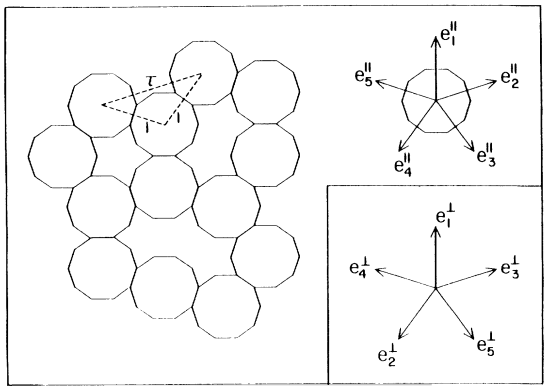

FIG. 1. Example of a decagon packing and projected lattice generators $\mathbf{e}_i^{\parallel}$. Inset: projected lattice generators $\mathbf{e}_i^{\perp}$.

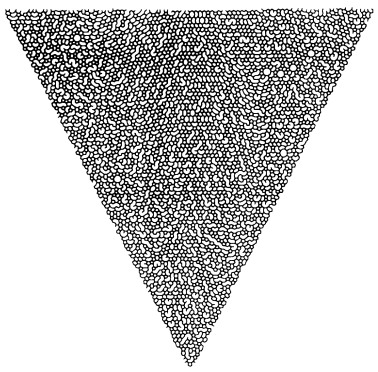

FIG. 2. Example of a grown decagon packing showing the network formed by nn bonds.

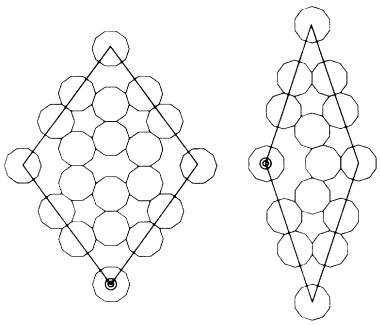

FIG. 3. Decoration of the 2D Penrose rhombi with decagons.

bonds and σ_τ for the nnn bonds. Both Lennard-Jones depths are the same. The original edge-sharing structure, or reference solid, is stabilized by the choice $\sigma_\tau/\sigma_1=\tau$. We are interested in the formation of strains (deformation of the reference solid) when σ_τ/σ_1 differs *infinitesimally* from τ. In this limit (*elastic regime*) the actual form of the potential is irrelevant since the harmonic behavior dominates the dynamics.

The constraints on the local geometry of our decagon packing still permit a large number of possible strain-free reference solids. We have studied two kinds of packings that can easily be distinguished by the behavior of the phason coordinate. The first is a quasiperiodic packing constructed by our decorating the 2D Penrose tiling of rhombi with decagons as shown in Fig. 3. The variation of $\mathbf{x}^\perp$ from decagon to decagon is shown in Fig. 4(a) by means of a vector proportional to $\mathbf{x}^\perp$, based at $\mathbf{x}^\parallel$ for each decagon with 4D coordinates $(\mathbf{x}^\parallel,\mathbf{x}^\perp)$. Although there are rapid local variations in $\mathbf{x}^\perp$, there are no systematic changes on long length scales. In fact, a necessary condition for quasiperiodic long-range order (of the reference solid) is simply that the $\mathbf{x}^\perp$ differences are bounded.

The second kind of decagon packing we have studied was generated by a growth algorithm. Our algorithm[5] is an extension of earlier aggregation models[6] which avoids the formation of 1D defects, or "tears," in the connectivity of the bond network. The growth geometry is a triangle with one edge moving at constant velocity v away from the opposite vertex. Using coordinates $\mathbf{x}^\parallel=(x,y)$, where y represents the growth direction, the interior of the triangle is given by $2|x|<y$, $0<y<y_0$. The growth nucleus is a single decagon placed at (0,0). High connectivity of the network is achieved by Metropolis annealing with a linear temperature field $T(y)=h(y-y_0)$. The growth velocity is established by the motion of the zero-temperature isotherm: $\dot{y}_0=v$. Each decagon-decagon bond, both nn with length 1 and nnn with length τ, is assigned a cohesive energy of -1. Growth and annealing occur in the region $T>0$ $(y>y_0)$. A single growth-thermalization process consists of the following two operations applied to one of the N decagons, say D_1, chosen at random from the region $y>y_0$. (1) A bond emanating from D_1 is chosen at random and if a decagon D' placed at the other end of the bond satisfies two properties it is added to the structure: (i) It is simultaneously bonded to at least one other decagon, say D_2, and (ii) the distance between D' and other decagons to which it is not bonded is greater than τ. (2) D_1 may be removed according to the Metropolis criterion: a random number r, uniform in (0,1) is chosen and if $r<\exp(-n_{\text{bond}}/T)$, then D_1 is removed. Here n_{bond} is the loss of cohesive energy given by the number of bonds removed when D_1 is removed and $T(y)$ is the local temperature. At the completion of each growth-thermalization process the zero-temperature isotherm is advanced according to $y_0\rightarrow y_0+v/N$.

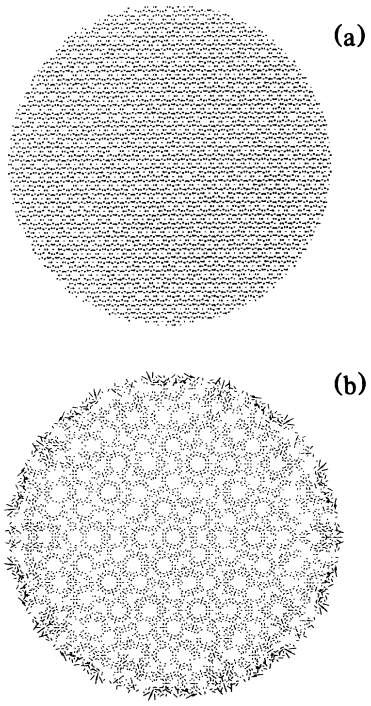

FIG. 4. (a) Phason field of the quasiperiodic packing; (b) fiftyfold magnification of the displacement field of the same packing.

We find that the parameter values $h=0.3$ and $v<0.001$ produce satisfactory networks without tears at the length scales considered here. Figure 5(a) shows the phason field of a circular region excised from the center of the triangular aggregate shown in Fig. 2. A striking feature of Fig. 5(a) is the long-wavelength variation in $\mathbf{x}^\perp$. This is especially remarkable in view of the uniformity of the growth process. The same feature has been observed in analogous simulations of a 3D icosahedral model.[7]

The rigid geometries of our two reference solids, the

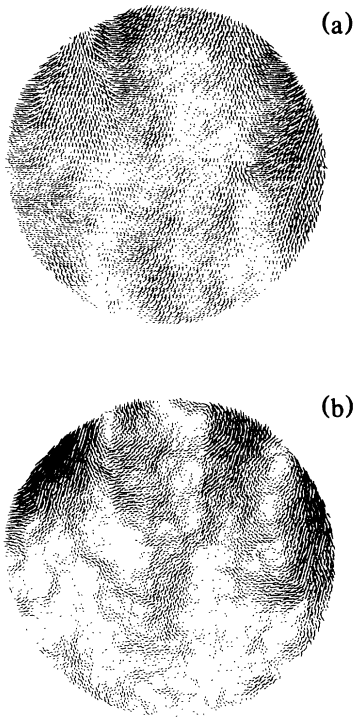

FIG. 5. (a) Phason field of a grown decagon packing; (b) tenfold magnification of the displacement field of the same packing.

quasiperiodic and grown decagon packings, correspond to a very nongeneric case of interparticle potentials. There are no symmetry principles that require that the ratio of the bond lengths is precisely τ or that the angles between bonds are precise multiples of 36°. Indeed, by imposing pair potentials with $\sigma_\tau/\sigma_1=\tau+\delta$, $\delta\neq 0$ these properties disappear. We have generated relaxed decagon packings for $\delta=0.1$ using a molecular-dynamics algorithm. This value of δ is small enough that the response (i.e., displacement field) scales linearly with δ. In the relaxed packings, not all the lengths of nn and nnn bonds are exactly at the minimum of their respective pairwise potentials.

A trivial consequence of modifying the potential is a uniform strain, e.g., isotropic contraction or expansion, of the unrelaxed packing. The uniform component of the strain was eliminated with the method of least squares. Let $\mathbf{x}_i$ ($\mathbf{x}_i''$) denote the unrelaxed (relaxed) positions of the ith decagon and $\mathbf{x}_i'$ the linear change applied to the unrelaxed position, i.e., $\mathbf{x}_i'=\mathbf{x}_i+\mathbf{A}\mathbf{x}_i+\mathbf{b}$. The uniform strain matrix $\mathbf{A}$ and translation vector $\mathbf{b}$ are determined by our minimizing the expression $\Delta^2=N^{-1}\sum_{i=1}^{N}(\mathbf{x}_i''-\mathbf{x}_i')^2$. Δ gives the root mean square (rms) displacement, i.e., the "random" strain when the trivial effects given by a linear transformation (rotation, expansion, translation, shear, etc.) have been eliminated.

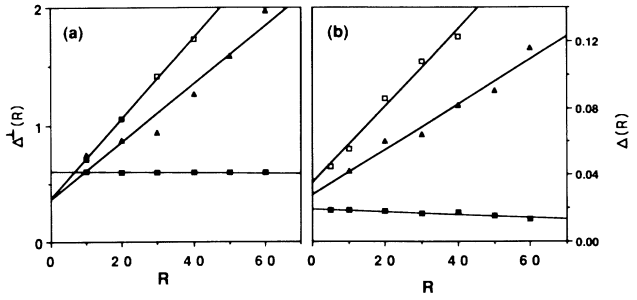

FIG. 6. Root mean square (a) phason and (b) ordinary displacements for the three decagon packings: quasiperiodic (■); grown, with $h=0.3$, $v=0.0015$ (▲); grown, with $h=0.3$, $v=0.0005$ (□). The data are plotted for circular packings with radius R.

The random-strain field of the quasiperiodic packing is shown in Fig. 4(b). Although the strain field has destroyed the bond-length–angle relationships of the rigid geometry, it is clear that the relaxed structure is still quasiperiodic. To see this we note that the strain field itself is quasiperiodic. This is evident from Fig. 4(b) and is easily explained since (i) each decagon displacement is determined by its environment (in the reference solid) and (ii) the set of similar environments forms a quasiperiodic pattern. Figure 4(b) also suggests that the decagon displacements (after subtraction of the uniform component) are bounded. We believe this is generally true for quasiperiodic structures, provided the forces are short ranged and the perturbation of the potential is sufficiently small.

The behavior of the strain fields of the grown decagon packings is quite different, as Fig. 5(b) shows. Again, we have subtracted the uniform component of the strain so that only the random component, associated with inhomogeneities, remains. There is clearly an accumulation of strain in that the large displacements away from the reference solid involve the coherent motion of many decagons. The growth of the rms displacement, $\Delta(R)$, with the radius R of the packing is shown in Fig. 6(b). In the quasiperiodic packing, ΔR quickly saturates as a function of R whereas each of the grown packings we have studied show a linear rise in $\Delta(R)$. A comparison of Figs. 5(a) and 5(b) suggests that the inhomogeneity responsible for the accumulation of strain in the grown packings is the long-wavelength variation of the phason coordinate. It is interesting that a plot of $\Delta^{\perp}(R)$, the rms phason displacement (with linear component subtracted) also shows a linear rise [Fig. 6(a)].

Several diffusion experiments[8,9] have established a linear growth of peak width with phason momentum $\mathbf{G}^{\perp}$ in quasicrystals. The apparent linear growth of $\Delta^{\perp}(R)$ with R in Fig. 6(a), seen also in the 3D icosahedral model,[7] is consistent with this behavior. Specifically, if $\mathbf{x}_i^{\parallel}$ is the position of the ith decagon in the reference solid and

$\delta\mathbf{x}_i$ its displacement after relaxation, then the scattering phase angle is given by[4]

$$\mathbf{G}^{\parallel}\cdot(\mathbf{x}_i^{\parallel}+\delta\mathbf{x}_i)=-\mathbf{G}^{\perp}\cdot\mathbf{x}_i^{\perp}+\mathbf{G}^{\parallel}\cdot\delta\mathbf{x}_i\ (\mathrm{mod}2\pi)\,. \quad (1)$$

Fluctuations in both $\mathbf{x}_i^{\perp}$ and $\delta\mathbf{x}_i$ lead to peak broadening. In particular, for peaks with $|\mathbf{G}^{\perp}|\gg|\mathbf{G}^{\parallel}|$ one considers a coherence radius R_c defined by $|\mathbf{G}^{\perp}|\Delta^{\perp}(R_c)\sim\pi$. From the behavior $\Delta^{\perp}(R)\sim aR$ shown in Fig. 6(a) one then obtains a peak broadening $\delta G\sim\pi/R_c\sim a|\mathbf{G}^{\perp}|$.

Systematic departures from linear $|\mathbf{G}^{\perp}|$ peak broadening have been noted for diffraction peaks in the opposite limit: $|\mathbf{G}^{\parallel}|\gg|\mathbf{G}^{\perp}|$. The interpolating form $\delta G^2=|a\mathbf{G}^{\perp}|^2+|b\mathbf{G}^{\parallel}|^2$ has been fitted to experimental data with some success[8,9] and Horn *et al.*[8] have argued that a $\mathbf{G}^{\parallel}$ term is a consequence of dislocations. The same argument given above, but applied to the rms fluctuations in $\delta\mathbf{x}_i$, leads to peak broadening of the form $\delta G\sim b|\mathbf{G}^{\parallel}|$, where now b comes from the behavior $\Delta(R)\sim bR$ shown in Fig. 6(b). Thus, our model, which is *free of dislocations,* reproduces the main features of peak broadening in quasicrystals. Experimentally, the ratio a/b is large.[9] Our results, where b depends linearly on δ, give a similarly large ratio, suggesting that the analog of δ in real quasicrystals is also small. Finally, we find (e.g., see Fig. 6) a dependence of strain accumulation on cooling rate, in that slow cooling induces larger strain. This surprising result is consistent with recent experiments in Ga-Mg-Zn.[10]

We acknowledge useful conversations with C. Henley. We are indebted to Eduardo Fradkin and the computing center of the Materials Research Laboratory of the University of Illinois for their invaluable assistance. This work has been supported in part by the NSF through Grants No. MRL-DMR-86-12860, No. PHY82-17853, supplemented by funds from NASA, and by DOE through Grant No. DE84-ER-45108.

[a]Permanent address: Dipartimento di Fisica, Universitá degli Studi di Trento, 38050 Povo (TN), Italy.

[b]Permanent address: AT&T Laboratories, Murray Hill, NJ 07974.

[1]J. C. Phillips, Phys. Today **35**, No. 2, 27 (1982).

[2]D. R. Nelson, Phys. Rev. B **28**, 5515 (1983); P. J. Steinhardt, D. R. Nelson, and M. Ronchetti, Phys. Rev. Lett. **47**, 1297 (1981), and Phys. Rev. B **28**, 784 (1983).

[3]Our definition of the word "quasicrystal" corresponds to the experimental one: A solid having a sharp but noncrystallographic diffraction pattern. In addition to quasiperiodic structures this definition includes random aggregation models such as the icosahedral glass.

[4]An excellent survey of the literature is given by C. L. Henley, Comments Condens. Mater. Phys. **13**, 59 (1987).

[5]V. Elsin, in *Proceedings of the Fifteenth International Colloquium on Group Theoretical Methods in Physics,* edited by R. Gilmore and D. H. Feng (World Scientific, Singapore, 1987), Vol. 1, p. 162.

[6]D. Schechtman and I. Blech, Metall. Trans. A **16**, 1005 (1985); P. W. Stephens and A. I. Goldman, Phys. Rev. Lett. **56**, 1168 (1986), and **57**, 2331 (1986).

[7]V. Elser, in *Proceedings of the NATO Advanced Research Workshop on New Theoretical Concepts in Physical Chemistry,* edited by A. Amann, L. S. Cederbaum, and W. Gans (Reidel, Dordrecht, 1988).

[8]P. M. Horn *et al.*, Phys. Rev. Lett. **57**, 1444 (1986); P. A. Heiney *et al.*, Science **238**, 660 (1987).

[9]D. Gratias *et al.*, to be published.

[10]W. Ohashi and F. Spaepen, Nature (London) **330**, 555 (1987); H. S. Chen *et al.*, Phys. Rev. B **38**, 1658 (1988).

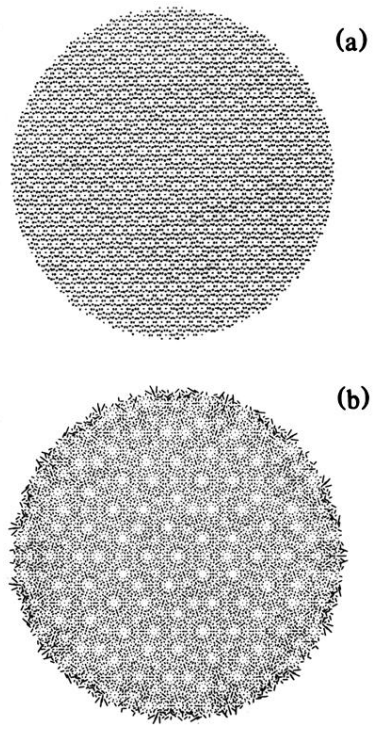

FIG. 4. (a) Phason field of the quasiperiodic packing; (b) fiftyfold magnification of the displacement field of the same packing.

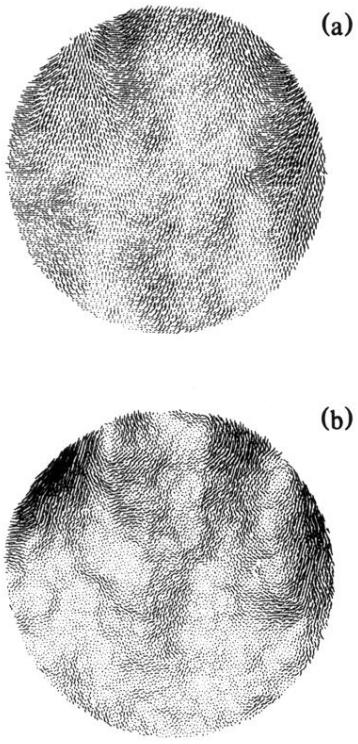

FIG. 5. (a) Phason field of a grown decagon packing; (b) tenfold magnification of the displacement field of the same packing.