Anomalous diffusion and conductivity in octagonal tiling models

B. Passaro
Dipartimento di Fisica, Universitá di Milano, Sezione di Milano, Istituto Nazionale di Física Nucleare, Via Celoria 16, 20133 Milano, Italy

C. Sire
Laboratoire de Physique Quantique, Université Paul Sabatier, 31062 Toulouse CEDEX, France

V. G. Benza
Dipartimento di Fisica, Universitá di Milano, Sezione di Milano, Istituto Nazionale di Física Nucleare, Via Celoria 16, 20133 Milano, Italy

(Received 7 July 1992)

We present numerical calculations of the quantum diffusion over an octagonal quasiperiodic tiling. We have studied a one-parameter family of Hamiltonians including the pure hopping case, the Laplacian, and a regime where atomic potentials prevail. We have found that unlimited diffusion occurs with anomalous exponents both in the hopping regime, where the spectrum has a band structure, and in the strong-coupling regime, where the spectrum has a Cantor structure. Upon introducing disorder in the lattice through phasonic fluctuations, the diffusion exponent increases in the pure hopping regime, while localization appears in the strong-coupling regime. The consequences on the conductivity of real quasicrystals are considered.

I. INTRODUCTION

The discovery of thermodynamically stable quasicrystals,[1] together with the possibility of producing samples with low density of defects, allows the exploration at the experimental level of the physical properties associated with quasiperiodicity, which have been the object of interest in the scientific community since long ago.

The low density of states (DOS) at the Fermi level could be at the origin of the anomalously high resistivity observed.[2] In fact on the basis of Hume-Rothery rules a pseudogap at the Fermi level was predicted,[3] subsequently confirmed by numerical simulations,[4] and finally observed[5] by means of x-ray absorption and emission experiments. Contrary to what was expected, it was also observed that pure samples are more resistive than disordered ones. The conductivity increases in a dramatic way as the temperature increases. It is then of fundamental interest to understand such behavior on theoretical grounds.

In spite of a large amount of work on one-dimensional quasiperiodic Hamiltonians, electronic properties of quasiperiodic tilings in two and three dimensions are unknown. In one dimension it is by now an established fact that the wave functions are neither extended nor localized, but decay with power law at large distances, and correspondingly the spectrum has a Cantor structure.

Some numerical works on finite clusters of two-dimensional (2D) Penrose tilings gave interesting information on the possible coexistence of localized and extended states but were unable to establish whether the spectrum has a band structure or is Cantor-like as in the one-dimensional case.[6] Progress on this question has recently been made by two of us[7] in studying the two-dimensional octagonal tiling. It was established without ambiguity that in the generic case the Lebesgue measure of the spectrum is finite with a finite number of gaps and that the wave functions may be extended on very large regions. There is no reason to think that such a feature is specific of the eightfold symmetry, but rather it is expected to hold in general, e.g., in 2D or 3D Penrose lattices. Work in this direction is in progress.

While former approaches dealing with pure point spectra did not allow one to locate the gaps, our approach was based on scaling analysis over a sequence of periodic tilings with increasingly large square unit cells.[8] Bloch boundary conditions allowed us to determine pure band spectra at each order.

In Ref. 7 it was also shown that when hopping dominates over atomic potentials there is level repulsion, as in quantum systems with a classically chaotic analog, while in the opposite regime there is level attraction. Level attraction was previously observed in one-dimensional quasiperiodic chains[9] and in the quasiperiodic Harper model at the metal-insulator transition.[10] As we will illustrate in the sequel, level attraction is an index of the occurrence of Cantor-like spectra, so that the global picture obtained in Ref. 7 involves a transition from a pure band spectrum to a Cantor-like spectrum in the regime of strong local coupling and weak hopping amplitude. In the following we extend our analysis to the degree of localization of the physical states in the various regimes, and study the quantum diffusion. We shall discuss the consequences of our results on the conductivity of real materials.

II. MODEL HAMILTONIAN

The octagonal quasicrystal (see Fig. 1) is generated by two elementary tiles, a square and a rhombus, and has six

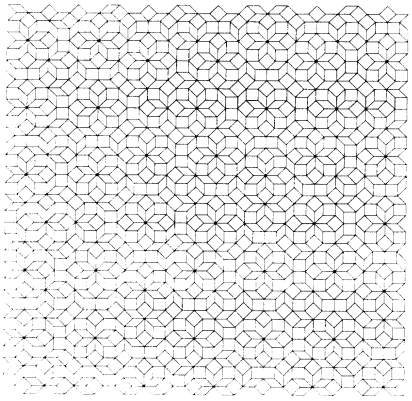

FIG. 1. Unit cell of the periodic approximant of the octagonal tiling with 1393 sites.

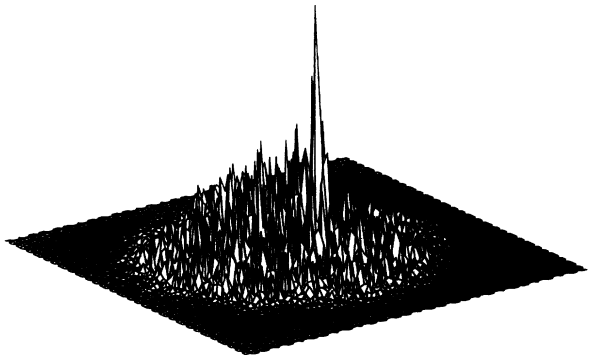

FIG. 2. Absolute value of the wave packet in the pure hopping regime $t=1$ at $\tau=12$.

types of vertices, respectively with coordination $z=3$, 4, 5, 6, 7, and 8. Each type of vertex occurs with a given frequency: $\nu_3=w$, $\nu_4=2w^2$, $\nu_5=2w^3$, $\nu_6=2w^4$, $\nu_7=w^5$, $\nu_8=w^4$ with $w=\sqrt{2}-1$. In our model the particle occupies the vertices of the tiling and is described by the parametric Hamiltonian

$$(H\psi)_i=t\sum_{j=1}^{z_i}\psi_j+(1-t)z_i\psi_i \quad (0<t\leq 1)\ , \tag{1}$$

where the sum extends over the nearest neighbors j of the site i. Notice that as t varies from 0 to 1, one goes from a strong-coupling regime to a pure hopping regime. When $t=\frac{1}{2}$, (1) coincides with the Laplacian over the lattice.

In Ref. 7 we obtained that the Lebesgue measure of the spectrum is certainly larger than zero when $t>0.2$, and that when $t_c<t$, ($t_c\approx 0.35$) there is level repulsion, while in the complementary regime there is level attraction. Due to finite-size effects, it was impossible to establish whether indeed the Lebesgue measure is zero when t is small enough. On qualitative grounds, one then expects extended states above t_c and possibly coexistence of extended states and critical states below t_c. The topology of the lattice also allows strictly localized (with compact support) energy eigenstates: this feature, first noticed in the Penrose tiling,[6] does not seem to be physically relevant. Indeed, it strongly depends on the form of the Hamiltonian.

We study the time evolution of a wave function initially concentrated at a given site (see Fig. 2), and in particular determine the spread of the wave packet for large times τ:

$$\Delta x(\tau)=\langle (x-\langle x\rangle)^2\rangle^{1/2}=D\tau^{\alpha}\ . \tag{2}$$

We average by sampling over different initial environments (typically of the order of 200) weighted by taking into account the relative frequencies of the various types of vertices, and truncate the regression when the wave function reaches a fixed distance from the boundary of the unit cell. We consider periodic approximants with 239, 1393, 8119, and 47 321 sites. We have computed the spread both by directly integrating in time and by diagonalizing the operator with periodic boundary conditions. The obtained exponents are given in Table I. The order of the approximants examined gave a satisfactory degree of convergence in the estimates.

III. HOPPING REGIME

When $t=1$ the spectrum reduces to a single band,[7] so that one would expect a ballistic diffusion: we obtained instead $\alpha=0.78$. A second quite unexpected feature is that $\Delta x(\tau)$ is strongly sensitive to initial conditions, more precisely, all plots referring to homologous (with equal z) initial sites cluster together, and the different clusters never intersect (see Fig. 3). For this reason we computed the exponents for the different ensembles. Notice that α

TABLE I. (a) Diffusion exponents α for the deterministic case. (b) Diffusion exponents α for the disordered case.

t	α	α_3	α_4	α_5	α_6	α_7	α_8
			(a)				
1.0 (app.5)	0.781	0.754	0.786	0.807	0.809	0.826	0.840
0.5 (app.5)	0.734	0.752	0.717	0.678	0.646	0.581	0.573
0.35 (app.4)	0.620	0.696	0.564	0.510	0.484	0.486	
0.1 (app.4)	0.359	0.529	0.179	0.047			
			(b)				
1.0 (app.5)	0.815	0.798	0.820	0.841	0.842		
0.5 (app.5)	0.628	0.656	0.615	0.602	0.603		

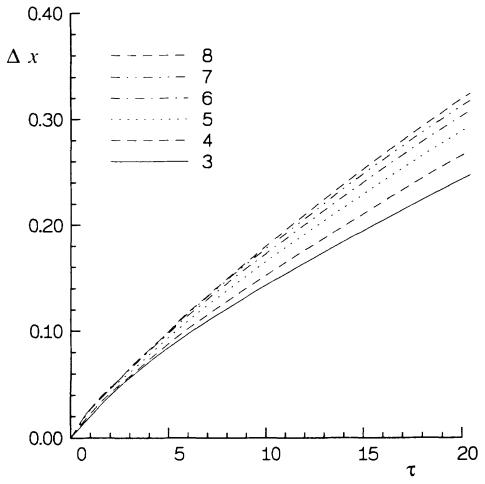

FIG. 3. Time evolution of Δx for initial conditions with coordination numbers $z=3,4,5,6,7,8$ ($t=1$).

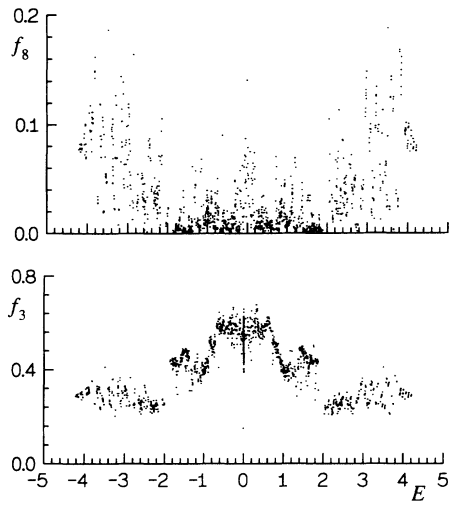

FIG. 5. Occupation probabilities $f_3(E)$ (down) and $f_8(E)$ (up) (see text).

increases with z. A partial explanation of this behavior can be given by noticing that the energy eigenfunctions at the band edges are in this case more extended than the corresponding ones at the band center, and furthermore that for energies around the band center the particle is more likely to stay on a small z site, while for energies at the band edges it is more likely to stay on a large z site. In order to convince oneself of the first property, one is led to examine the inverse participation ratio (IPR) (Ref. 11) of the eigenfunction ψ_E: $I(E)=\sum_i |\psi_{i,E}|^4$.

We recall that for a lattice having N sites, if ψ is a plane wave $I \approx 1/N$, if ψ is localized at a site $I \approx 1$, so that the larger the I the stronger the localization. Here, we have found that the eigenfunctions at the band edges are more extended (see Fig. 4).

The second property emerges from examining the probability $f_z(E)$ of the sublattice with coordination z being occupied at energy E. In Fig. 5, notice that f_3 is concentrated at the band center while f_8 is concentrated at the band edges. One can conclude that a state concentrated at a large z site predominantly selects the faster high $|E|$ eigenstates. We also studied the effect of phasonic disorder, generated upon flipping within the hexagons the position of the squares (see Fig. 6). The IPR in this case indicates that the relative weight of the "less extended" portion of the eigenfunctions is reduced. A clearer signature of phasonic disorder comes from the integrated density of states $N(E)$ (see Fig. 7). We believe that the irregular behavior of $N(E)$ in the deterministic case reveals a singular continuous spectrum; several pseudogaps can be found, notice in particular the large ones at $|E| \approx 2$. The discontinuity at $E=0$ comes from localized eigenstates. The phasons produce a smoothing of $N(E)$, so that the pseudogaps practically disappear: this implies a tendency towards a faster diffusion. Correspondingly, we obtain higher diffusion exponents (see Table I). This confirms on qualitative grounds the experimental results on resistivity, although the observed effect seems to be a stronger one. Of course, other disordering mechanisms should be taken into account in a more detailed modeling.

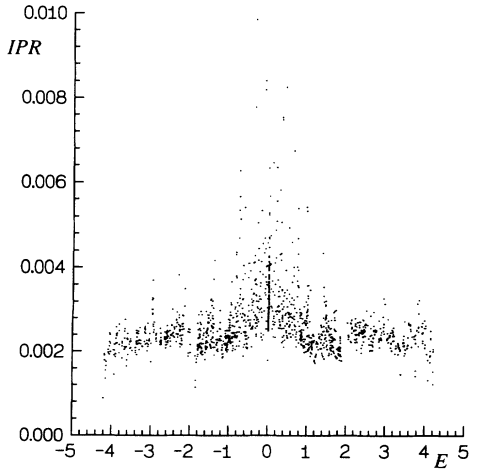

FIG. 4. Inverse participation ratio for $t=1$.

IV. STRONG-COUPLING REGIME

When t is small on qualitative grounds the standard arguments of Anderson localization hold: one actually is dealing with perturbation of the $E=z$ degenerate levels. Tunneling between homologous sites placed at a given distance will produce a splitting of some order; the further the sites, the smaller the splitting. The underlying scale invariance of the lattice is expected to induce a

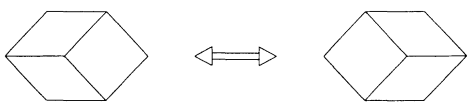

FIG. 6. Elementary operation including disorder in the tiling.

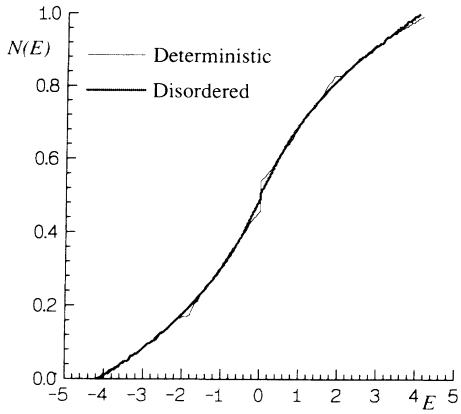

FIG. 7. Integrated density of states ($t=1$) for deterministic and disordered tiling.

hierarchy in the splittings as can be found from a renormalization group analysis.[14] This property implies a time evolution regulated over various time scales. More clearly, we observed that when starting from high z sites the wave function is at later times concentrated practically only on homologous sites and that diffusion appears as an intermittent process in the form of tunneling between metastable stationary states. Each such state is concentrated over a given pattern of homologous sites. Again one can determine a diffusion exponent for the various "bands" of the spectrum, with the result that here, opposite to the previous case, α decreases with z. This fact is an obvious consequence of the larger relative distance of the high z sites. The exception of the $z=7$ sites, more diffusive than the $z=8$ sites in spite of being rarer, can be easily explained by examining the second order correction in t. Even if we are not able to give a direct proof that when t is close to zero the spectrum is fractal, nonetheless various arguments can be put forward in favor of this hypothesis.

We first mention an extension of the argument given in Ref. 12 that allows us to relate the information dimension β ($\beta \leq d_H =$ Hausdorff dimension) of the spectrum with the diffusion exponent[13] in terms of a lower bound, which in two dimensions is

$$\Delta x(\tau) \geq \left[\frac{\tau^{\beta}}{\log \tau} \right]^{1/2} . \tag{3}$$

From (3) it follows that if $\alpha < 0.5$ the spectrum has a dimension smaller than one. In fact this is the case for each "band" at $t=0.1$ (see Table I). Presumably, the spectrum is a multifractal, each "band" having a different dimension. As t increases α increases, and correspondingly at some critical value t_c' the dimension reaches the value of $d_H=1$. From (3), one finds that $\beta \leq 2\alpha$. Note that in our case the inequality is found to be strict (e.g., $t=0.1$, $\beta \approx 0.5$, and $\alpha \approx 0.36$), contrary to the 1D case where one has $\beta=\alpha$.

A second, independent check of this picture comes from level statistics, where as was already stated, a transition between a level attraction regime and a level repulsion regime occurs at $t_c \approx 0.35$. If the number $P(s)$ of level spacings larger than s behaves as $s^{-\alpha}$ when $s \to 0$, one can conclude heuristically that a portion of the spectrum has dimension $d_H=\alpha$. In fact, if γ is the dimension, such portion is covered by $N_s=(1/s)^{\gamma}$ intervals with spacing s. N_s also counts the number of level spacings smaller than s, so that the number of spacings of order s diverges as $(1/s)^{\gamma+1}$. This implies $P(s)=(1/s)^{\gamma}$. In general we will have $t_c' < t_c$, in that when $t > t_c$ the spectrum has no fractal portions. We have also explored the effect of phasonic disorder at $t=0.1$. Upon destroying the recurrences in the tiling related with its scale invariance, the effects of resonance of homologous sites become frustrated. We verified this effect and the result is localization. Work is in progress on the disordered tiling in order to locate the transition to anomalous diffusion.

V. CONDUCTIVITY

In this section we will summarize our results and make some comparison with other works on the conductivity properties of quasicrystals. Octagonal tilings are characterized by anomalous diffusion with nonuniversal exponents: we believe this to be true in general for 2D quasiperiodic tilings with a generic Hamiltonian. Propagation is strongly influenced by quasiperiodicity in the strong and intermediate coupling regimes, in the form of tunneling between metastable stationary states, concentrated over clusters of homologous atoms. In the regime of weak local coupling there is pure band spectrum, but again anomalous diffusion occurs: this is compatible with (3), while in the one-dimensional case the diffusion exponent is bounded from below by the dimension of the spectrum, so that this anomaly never occurs.

In the strong-coupling regime the phasonic disorder induces localization; in the hopping regime, it enhances the propagation. The latter behavior confirms the experiments on conductivity, where apparently a stronger effect was observed. In terms of our parametric Hamiltonian everything goes as if disorder would produce a t_{eff} larger than the parameter t associated with the deterministic tiling: this implies stronger local potentials for defect-free materials.

In real materials (say Al-Cu-Fe), besides the aforementioned increase of the conductivity σ with disorder, a strong enhancement of σ with temperature is also observed. It seems that the naive scheme where σ is simply proportional to the DOS at the Fermi energy $[\rho(E_F)]$ is not consistent with this behavior. One could try to explain the fact that σ has stronger variation than $\rho(E_F)$ by the nonsphericity of the pseudo-Fermi surface[15] or by a Mott-like behavior $\sigma \sim \rho(E_F)^2$.[16] Even if these effects should be considered, on the basis of the results presented here we propose an alternative explanation.

In a more realistic electronic model for quasicrystals, the Al-Cu atoms are described by an effective s band, the transition atoms (Fe here) by a narrow d band, with some hybridization[17] between them. The on-site potential on transition atoms is chosen such that the narrow d band falls close to E_F of the unperturbed effective s band. Thus it may be quite different from the potential associated with the s band. Of course $\rho(E_F)$ in such models can

be set quite low but still not low enough to explain the weak conductivity observed, except by considering unphysical very strong couplings.

We propose that the states around the Fermi level are poorly conducting ones (because the density of transition atoms is low) and will resemble the states already described in our 2D model, localized on (many) similar regions ("quasilocalized" in the following). However, they are *not* localized in the usual sense, and will not contribute to the specific heat like localized states would do. Thus, even if $\rho(E_F)$ is not so low, conductivity might be much smaller than expected. Upon increasing the temperature, conduction will be allowed by these states through inelastic scattering. Works are in progress to test the conjecture of "quasilocalization" of states near E_F for a realistic 3D model of Al-Cu-Fe.[18]

ACKNOWLEDGMENTS

C. S. is grateful to the Laboratoire d'Etudes des Propriétés Electroniques des Solides, Centre National de Recherches Scientifiques (LEPES) group in Grenoble for their kind hospitality. C. S. particularly thanks D. Mayou, G. Trambly, C. Berger, N. W. Ashcroft, and S. Burkov for fruitful discussions.

[1]A. P. Tsai, A. Inoue, and T. Masumoto, Jpn. J. Appl. Phys. **26**, L1505 (1987).
[2]T. Klein, C. Berger, D. Mayou, and F. Cyrot-Lackmann, Phys. Rev. Lett. **66**, 2907 (1991).
[3]J. Friedel, Helv. Phys. Acta **61**, 538 (1988); A. P. Smith and N. W. Ashcroft, Phys. Rev. Lett. **59**, 1365 (1987).
[4]T. Fujiwara and T. Yokokawa, Phys. Rev. Lett. **66**, 333 (1991).
[5]E. Belin, J. Kojrisk, A. Sadoc, A. Traverse, M. Harmelin, C. Berger, and J. M. Dubois, J. Phys. Condens. Matter **4**, 1057 (1991).
[6]M. Kohmoto and B. Sutherland, Phys. Rev. B **34**, 3849 (1986).
[7]V. G. Benza and C. Sire, Phys. Rev. B **44**, 10 343 (1991).
[8]M. Duneau, R. Mosseri, and C. Oguey, J. Phys. A **22**, 4549 (1989).
[9]K. Machida and M. Fujita, Phys. Rev. B **34**, 7367 (1986).
[10]T. Geisel, R. Ketzmerick, and G. Petschel, Phys. Rev. Lett. **66**, 1651 (1991).
[11]R. J. Bell and P. Dean, Discuss. Faraday Soc. **50**, 55 (1970).
[12]B. V. Chirikov, F. M. Izrailev, and D. L. Shepelyanski, Sov. Sci. Rev. C **2**, 209 (1981).
[13]I. Guarneri, Europhys. Lett. **10**, 95 (1989).
[14]C. Sire and J. Bellissard, Europhys. Lett. **11**, 439 (1990).
[15]N. W. Ashcroft and S. Burkov (unpublished).
[16]C. Scheidt, K. Wang, and P. Garoche (unpublished).
[17]G. Trambly, D. Nguyen Manh, and D. Mayou (unpublished).
[18]D. Mayou and C. Sire (unpublished).

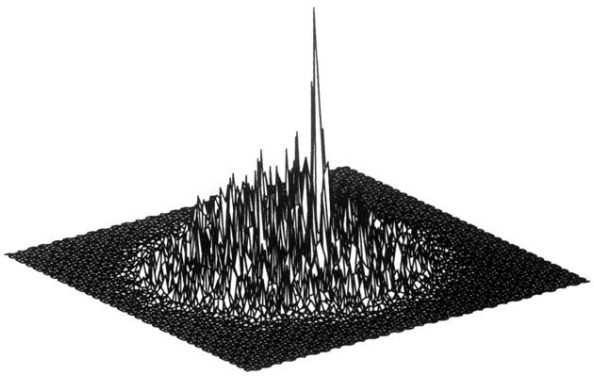

FIG. 2. Absolute value of the wave packet in the pure hopping regime $t=1$ at $\tau=12$.