Limited conductivity in an octagonal quasicrystal

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Conductivity of an *s*-band model on the octagonal tiling is studied. Using perturbation theory it is shown that there is only one channel available for electronic transport, when the kinetic term in the Hamiltonian is small. This feature is independent of the size of the approximant and reveals a microscopic mechanism that is capable of severely limiting conduction processes in a quasicrystal. Numerical evidence indicates that the same result is valid even in case of realistic kinetic energies at some values of the Fermi energy.

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I. INTRODUCTION

The discovery[1] of quasicrystals (QC's) in 1984 opened a new research area in condensed-matter physics. The interplay between quasiperiodic structure and physical properties is a central theme in this field. This paper focuses on the question why QC's, with structures intermediate between those of lattice-periodic crystals and amorphous materials, often have a conductivity that is an order of magnitude lower than that of nearby amorphous phases. A typical example is icosahedral (*i*)-AlCuFe. In a sample of high structural quality the conductivity at 4 K $\sigma_{4\ \mathrm{K}}$ can be as low as 90 $\Omega^{-1}\ \mathrm{cm}^{-1}$.[2] The conductivity is higher at higher temperatures and in samples of lower structural quality, i.e., with more disorder.[2] To compare: An amorphous phase of AlCuFe with atomic composition similar to the *i* phase has $\sigma_{4\ \mathrm{K}}=712\ \Omega^{-1}\ \mathrm{cm}^{-1}$,[3] liquid Fe has $\sigma_{1809\ \mathrm{K}}=7215\ \Omega^{-1}\ \mathrm{cm}^{-1}$,[4] in liquid Al, $\sigma_{933\ \mathrm{K}}=4.1\times10^{5}\ \Omega^{-1}\ \mathrm{cm}^{-1}$,[4] and face-centered cubic Al has $\sigma_{77\ \mathrm{K}}=3\times10^{6}\ \Omega^{-1}\ \mathrm{cm}^{-1}$.[5] So, in contrast to the structure, the conductivity of a QC is not intermediate between that of lattice-periodic crystals and amorphous materials. The origin of the anomalously low value of the conductivity of QC's still lacks a rigid microscopic explanation.

Considering the many theoretical papers that have been devoted to this problem, there is one obvious red line that illustrates the fact that the anomalously low conductivity in QC's *is* a surprise, and, in addition, makes clear why this problem has defied microscopic understanding so far: Electron diffusion in a generic quasiperiodic environment is superdiffusive. This has been shown for many quasiperiodic monatomic *s*-band tiling models, in particular, for the octagonal tiling (OT),[6,7] the two-dimensional (2D) Penrose tiling (PT),[8,9] the 2D generalized Rauzy tiling (GRT),[10,11] the 3D PT,[12] and the 3D GRT,[10] and, recently, also for a hypothetical decagonal (*d*)-Al phase.[13] Superdiffusive transport implicates infinite zero-temperature conductivity.[14] In contrast, in liquid metals elastic electron scattering due to disorder leads to diffusive transport, which corresponds to a finite value of the conductivity.[15,16] So, calculations have led to the intuitive result that, in general, electron diffusion in a quasiperiodic substrate is intermediate between ballistic diffusion in a lattice-periodic crystal and diffusive transport in a disordered medium, which reflects the intermediateness in structure, but is in clear contrast with experimental findings. In other words, the quasiperiodicity of a QC is not sufficient to explain its low conductivity.

Nevertheless, *ab initio* calculations have given evidence that elastic electron scattering *is* responsible for low QC conductivity. When Fujiwara *et al.*[17] studied a series of approximants to *d*-AlCuCo, they found slowly decreasing diffusion constants for some wave functions as a function of system size. Solbrig *et al.*[18] found that coherence on length scales larger than 12 Å, the size of a unit cell of a 1/1 approximant, is relevant for electronic transport in *i*-AlMn. In contrast, Krajčí and Hafner[19] found an increased conductivity for *i*-AlPdRe when they went from a 1/1 to a 2/1 approximant. They noted that *ab initio* calculations are extremely sensitive to, e.g., the modeled chemical composition. A detailed microscopic understanding of low QC conductivity was not provided in any of the above-mentioned papers. Some of the difficulties met in *ab initio* calculations are circumvented by using tiling models, which may thus be the easier way of providing a first step in the analysis of low QC conductivity.

In this paper I study a nontrivial 2D quasiperiodic tiling model. By means of perturbation theory I derive an effective Hamiltonian for the low kinetic energy limit. I show that in this limit the tiling splits up in clusters and chains. Only open chains contribute to the conductance. This feature provides an upper bound for the latter, which is shown to stay constant with increasing size of the approximants. I present numerical evidence, to the effect that this result is not restricted to the low kinetic energy limit. Even when the kinetic and potential terms in the Hamiltonian are of comparable magnitude, the conductance is dominated by open chains for some values of the Fermi energy. Here a clear connection is made between the real-space structure of a QC and its conductivity.

II. MODEL

The model is based on the OT. In order to obtain the properties of the perfect quasiperiodic tiling a series of approximants[20] with increasing unit-cell size is studied. By analyzing how the conductance scales in this series of approximants, conclusions about the conductivity of the infinite tiling can be drawn. In the electronic model there is one basis state on each vertex of the tiling. These states can be thought of as atomic *s* states. The tight-binding Hamiltonian is

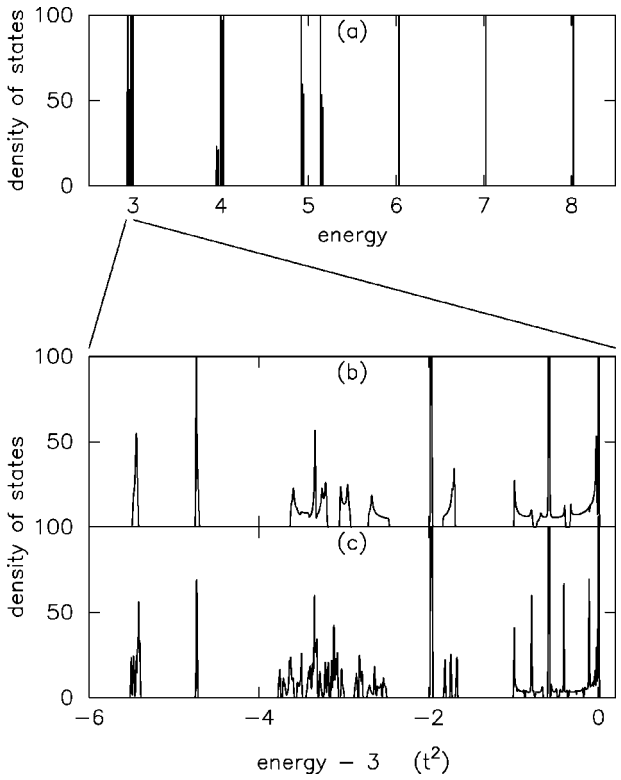

FIG. 1. (a) DOS of the OT with low kinetic energy, $t=0.11$. It consists of six narrow bands. (b) The substructure of the band at $E\approx 3$ has 0D dispersionless features and typical 1D Van Hove singularities. (a) and (b) were obtained for an approximant with 41 vertices per unit cell, (c) with 239 vertices. In the larger approximant additional gaps open up.

$$H=\sum_{\langle ij\rangle}|i\rangle t\langle j|+\sum_{i}|i\rangle n_i\langle i|, \tag{1}$$

with the first sum over all nearest neighbors, i.e., all pairs of vertices that are connected by an edge of the tiling, and n_i the coordination number of site *i*. t is a parameter that determines the relative weight of the first (kinetic) term of the Hamiltonian relative to the second (potential) term. Without the second term Eq. (1) would describe a monatomic *s*-band model. Distinguishing the on-site energies of the sites of the OT is a simple way of incorporating the effect of different kinds of atoms into the tiling model. As mentioned in the Introduction, without the second term transport in the OT is superdiffusive.[6,7]

The same model has been studied before.[6,7,21–24] When the hopping parameter t is small, the electronic density of states (DOS) consists of six narrow bands according to the possible numbers of nearest neighbors of the individual sites.[6,22] This is illustrated in Fig. 1(a). In this paper I study the band with $E\approx 3$. In Fig. 1(b) the substructure of this band is shown. There are dispersionless (0D) features and typical 1D Van Hove singularities. The bands between $E-3=-t^2$ and $E-3=0$ split up in subbands[6] when going to a higher approximant [Fig. 1(c)] in a similar way as, for example, the bands of approximants to the Fibonacci chain.[25] It has been noted that the substructure of each main band of Fig. 1(a) can in principle be described by perturbation theory.[6] Indeed, I found that the perturbational analysis in Sec. III A of this paper explains all the essential features (1D and 0D) in the DOSs of Figs. 1(b) and 1(c). The main benefit of the perturbation approach, however, lies in the insight it offers into the conductivity of the OT.

III. CONDUCTIVITY

A. Low kinetic energy limit

As was already mentioned in the previous section, the low kinetic energy limit of Eq. (1), i.e., $t\to 0$, can be described by degenerate perturbation theory.[6] Following Ref. 26, the Hamiltonian is written as H_0+H_1, where H_0 is the potential part, i.e., $\Sigma_i|i\rangle n_i\langle i|$, and H_1 is the relatively weak part that contains all hopping terms, i.e., $\Sigma_{\langle ij\rangle}|i\rangle t\langle j|$. It is useful to define the projection operators Q and $P=1-Q$, where Q projects onto all sites with coordination number three. The effective Hamiltonian in the subspace of all threefold coordinated sites is then to second order in t given by[26]

$$H_{\rm eff}=QH_0Q+QH_1Q+QH_1P\frac{1}{E-H_0}PH_1Q. \tag{2}$$

The first term of this equation is the on-site energy of the degenerate sites of interest, viz., three. As will be shown two threefold coordinated sites are never nearest neighbors of each other. Therefore, the second term in Eq. (2) equals zero. The third term leads to effective potentials and bonds proportional to t^2. For this reason I expressed the energy axis of Figs. 1(b) and 1(c) in units of t^2. By approximating $E\approx 3$ in Eq. (2), the effective Hamiltonian becomes energy independent.

By means of the cut-and-project method or with help of the inflation rules[27] for the OT it is readily verified that each threefold coordinated site is a nearest neighbor of either a sixfold, a sevenfold, or an eightfold coordinated site. These three possibilities are illustrated in Figs. 2(a)–2(c). From Figs. 2(a)–2(c) it is clear that two threefold coordinated sites are never in nearest neighbor positions, but they may be second nearest neighbors of each other. The environments of the sites with coordination number three of Figs. 2(a)–2(c) are fixed up to a sufficiently large distance, so that all effective second nearest neighbor bonds as well as the on-site energies are unique for the patches shown. For the sake of completeness, omitting the factors t^2, the effective potential of sites *A* and *B* equals $-11/6$, of sites *C* and *G* it equals $-7/4$, of sites *D*, *E*, and *F* it is $-9/4$, and of all sites of Fig. 2(c) it equals $-11/5$. The effective hoppings over the short diagonals of the rhombuses of Figs. 2(a), 2(b), and 2(c) equal $-4/3$, $-5/4$, and $-6/5$, respectively. The other bonds within the clusters of Figs. 2(b) and 2(c) are $-1/4$ and $-1/5$. The bonds that point outward of the clusters of Figs. 2(a) and 2(b) have strengths $-1/2$. Like the potentials all bonds are in units of t^2.

Via their outward pointing bonds the clusters of Figs. 2(a) and 2(b) form chains. The clusters of Fig. 2(c) remain isolated. Figure 2(d) shows the unit cell of an approximant to the OT. There is one closed and one open chain. If the cluster

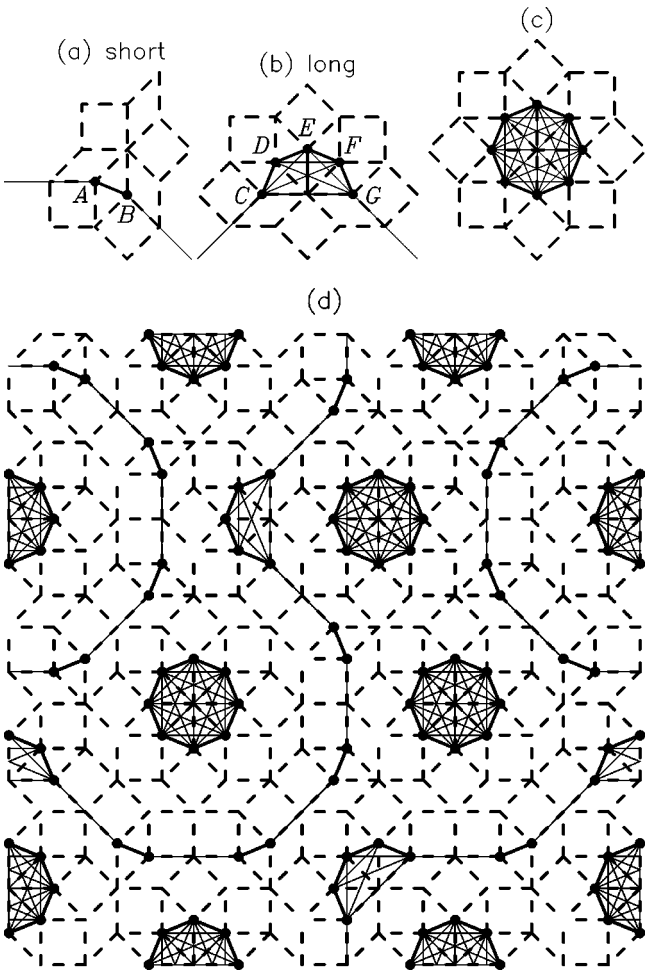

FIG. 2. Dashed lines indicate square and rhombus tiles of the OT. (a)–(c) show the possible environments of sites with coordination number three (dots). Solid lines are effective bonds in the low kinetic energy limit. In this limit, (a) and (b) form chains. (d) is the unit cell of an approximant with 239 sites. There is one closed and one open chain. The effective Hamiltonian of the open chain reproduces the 1D features in the DOS of Fig. 1(c).

of Fig. 2(a) is called short (S) and the one of Fig. 2(b) long (L), then the open chain of Fig. 2(d) is $SSSSLSLSL$. This chain is responsible for the 1D features in the DOS of Fig. 1(c). States on the closed chain and on the isolated clusters are dispersionless to second order in t, thus giving rise to the 0D features in Fig. 1(c). Larger approximants can be made by successive application of the inflation rules[27] of the OT. In each step the linear size of the unit cell increases by a factor of approximately $\sqrt{2}+1$. In this process isolated clusters and closed chains are inflated to isolated clusters and a larger number of closed chains. Some new isolated clusters appear. The open chain goes into an open chain three times longer according to the inflation rules

$$S \rightarrow SL, \tag{3a}$$

$$L \rightarrow SSSSL. \tag{3b}$$

These rules can be derived from the inflation rules[27] for the OT. In every inflation step the open chain becomes longer by a factor of three. It becomes more meandering. Therefore it has a fractal dimension $D=\log(3)/\log(\sqrt{2}+1)\approx 1.2465$. The substitution rules of Eq. (3) define an aperiodic chain. The most important result is that there is only one open chain in the unit cell of each approximant.

Since only one channel, viz., the open chain, is available for transport in the low kinetic energy limit, it follows immediately that the conductance of a unit cell of an approximant to the OT is at most e^2/h. This upper bound does not depend on the size of the approximant. Assuming that this bound is realized in all approximants, and that the thickness of the OT equals $l=1$ Å, I arrive at $\sigma=g/l\approx 3.87\times 10^3\ \Omega^{-1}\ \mathrm{cm}^{-1}$, which does not depend on the size of the unit cell. This value can of course not be taken seriously, since it was not derived from a realistic atomic model. The point is that the analysis of a quasiperiodic structure has proven its low conductivity and simultaneously provided considerable insight in the physical transport mechanism.

B. Realistic values of the kinetic energy

In order to study the effect of higher kinetic energy, I have used Landauer's formula. The procedure is analogous to the one in Ref. 9. In the unit cell of an approximant a set of parallel bonds is cut. The loose ends are connected to semi-infinite leads consisting of square tiles. In the perpendicular direction periodic boundary conditions are applied.[12] The results are averaged over a sufficient number of **k** points, so that full convergence with respect to the perpendicular extension of the system is reached. All sites in the leads have on-site energy three. In order to minimize the impedance misfit between the octagonal cell and the leads, the hoppings in the leads are chosen equal to t^2. Hoppings connecting the leads and the octagonal cell equal t^2 when they connect a site with coordination number three, and t otherwise. The conductance in this geometry is then evaluated as a function of the Fermi energy E_F with a Green's function method.[9,12] I have studied two cells, with 239 and 8119 sites.

For very low t (0.001) I found that the conductance is less than e^2/h for all values of the Fermi energy. When I increased t to 0.25, I saw that most values of the Fermi energy give an increased conductance. In a small part of the spectrum, however, the conductance had hardly changed. The calculated conductance in this energy range is shown in Fig. 3. It is seen that even for $t=0.67$ most features of Fig. 3(a) can still be recognized in the energy range $E_F-3\gtrsim -0.3\ t^2$. This result is quite unexpected, for in this case the kinetic and potential terms in the Hamiltonian are of comparable magnitude. $t=1$ gives an increased conductance for Fermi energies through the whole energy range shown, in agreement with the expectation that the above-derived perturbation theoretical result does not hold when the expansion parameter $t=1$. The fluctuations in Figs. 3(a)–3(c) are due to impedance mismatches between the leads and the octagonal cell. In general, each narrow band of Fig. 1(c) gives rise to one maximum. In higher approximants there are more bands and the conductance becomes a more strongly fluctuating function. Figure 3 provides strong numerical evidence that the remarkable low conductivity established for the low

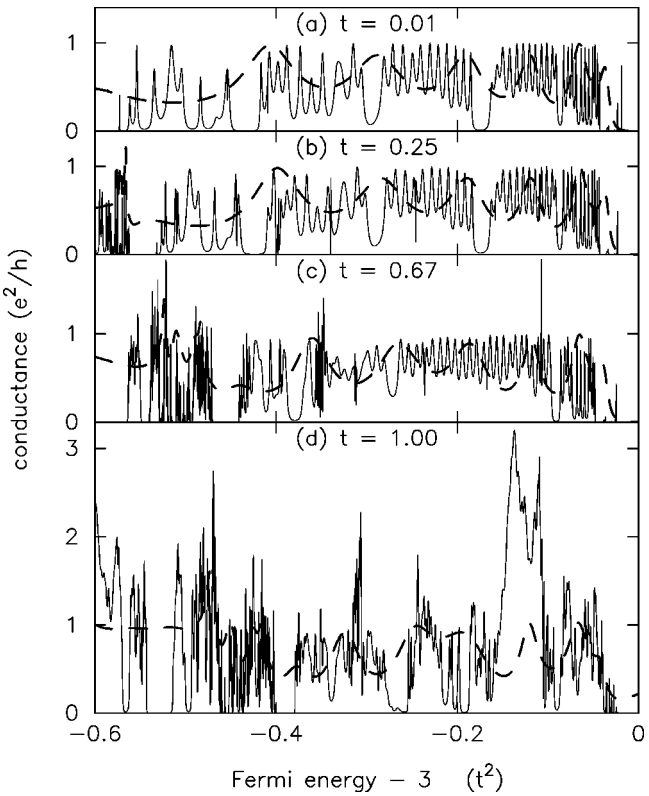

FIG. 3. Conductance of two cells of the OT with, respectively, 239 (dashed lines) and 8119 sites (solid lines). (a) When the kinetic energy is low, only the open chain of Fig. 2 can conduct. As a consequence, the conductance is at most one natural unit, i.e., e^2/h, independent of the approximant size. (b) For $t=0.25$ and (c) 0.67 this upper bound is not exceeded for some values of the Fermi energy. (d) For $t=1$, the conductance increases with increasing approximant size.

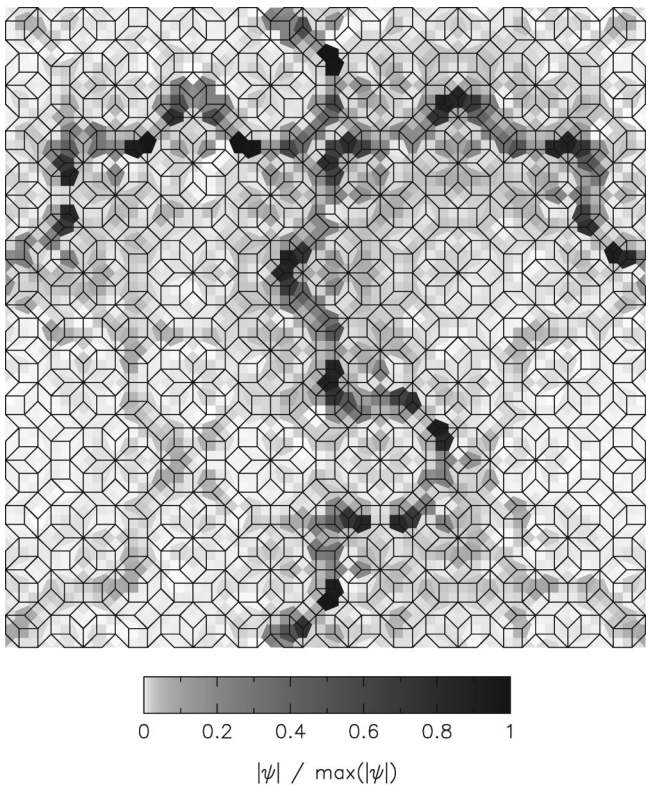

FIG. 4. Wave function on an approximant to the OT with 1393 vertices per unit cell. Antiperiodic boundary conditions apply. The energy of this state equals $3-0.19425\ t^2$, $t=0.67$. The wave function is localized on an open chain. This confirms the result of Fig. 3, that, for some energies, $t=0.67$ gives results that are not qualitatively different from those obtained with lower t.

kinetic energy limit survives when the hopping has the more realistic value $t=0.67$, at least for some values of the Fermi energy.

IV. DISCUSSION

By looking at the energy eigenfunctions of the Hamiltonian (1) I have obtained an independent confirmation of the surprising result that the description of the OT in terms of clusters and chains as given by degenerate perturbation theory (Sec. III A) is still valid for realistic values of the kinetic energy, e.g., $t=0.67$. Figure 4 shows a wave function with $E\approx 3-0.2\ t^2$, $t=0.67$. It was obtained by direct diagonalization[28] of the Hamiltonian (1). 91% of the weight of $|\psi|^2$ is on the open chain. This shows that, at the given energy and t, the effective Hamiltonian, Eq. (2), accurately describes the OT. I have also calculated wave functions for $t=1$. They are more delocalized, again indicating a breakdown of the perturbation theory for this value of t. Figure 4 illustrates the remarkable feature that the transport in the OT is locally anisotropic. Roche *et al.*[29] showed that the assumption of subdiffusive electron transport in QC's can qualitatively explain the anomalously low value of the conductivity. It was suggested that the subdiffusive behavior is a consequence of critical states in a QC. Such states are intermediate between exponentially localized states and extended states. They have a power-law envelope. It is expected that the amplitude of such a wave function falls off in roughly the same way in all directions. The mechanism described in this paper is locally anisotropic and should in that aspect be distinguished from the one of Roche *et al.*[29]

V. CONCLUSION

I have shown that in the OT electronic transport is dominated by a chain of atoms that meanders around the main clusters of the tiling. As a consequence conductivity is limited. I have given numerical evidence that for some values of the Fermi energy ($E_F-3\gtrsim -0.3\ t^2$) this is still true for realistic kinetic energies ($t=0.67$). It should be stressed that this paper does not intend to make a prediction about the particular class of QC's that have octagonal symmetry. The main result of this paper is a mechanism that is capable of limiting conductivity in a QC. When the quasiperiodicity is limited to two dimensions, as it is in the OT and in *d*-QC's, a fixed number of open channels independent of the approximant size gives an upper bound on the conductivity. For *i*-QC's, which are 3D, the number of open channels may grow proportional to the linear size of the approximants. The main challenge for the future is to find out whether this type of mechanism applies to real QC's.

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