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RESEARCH ARTICLE

Breakdown of the semi-classical conduction theory in approximants of the octagonal tiling

G. Trambly de Laissardière,^{†*} C. Oguey[†] and D. Mayou[‡]

[†] Laboratoire de Physique Théorique et Modélisation, CNRS UMR 8089, Université de Cergy-Pontoise, 2 av. A. Chauvin, 95302 Cergy-Pontoise, France

[‡] Institut Néel, CNRS, Université Joseph Fourier, Bât D, 25 av. des Martyrs, B.P. 166, 38042 Grenoble Cedex 9, France

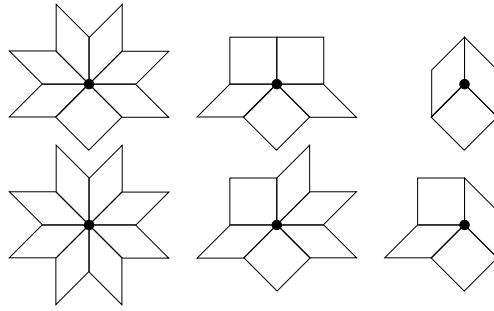
(August 2010)

We present numerical calculations of quantum transport in perfect octagonal approximants. These calculations include a Boltzmann (intra-band) contribution and a non-Boltzmann (inter-band) contribution. When the unit cell size of the approximant increases, the magnitude of Boltzmann terms decreases, whereas the magnitude of non-Boltzmann terms increases. It shows that, in large approximants, the non-Boltzmann contributions should dominate the transport properties of electrons. This confirms the break-down of the Bloch-Boltzmann theory to understand the transport properties in approximants with very large unit cells, and then in quasicrystals, as found in actual Al-based approximants.

1. Introduction

Since the discovery of Shechtman et al. [1] many experimental investigations have indicated that the conduction properties of several stable quasicrystals (AlCuFe, AlPdMn, AlPdRe...) are at the opposite of those of good metals [2, 3]. It appears also that the medium range order, over one or a few nanometres, is the real length scale that determines conductivity. In particular, the role of transition elements enhancing localisation has been often studied [4–11]. There is now strong evidence that these non standard properties result from a new type of break-down of the semi-classical Bloch-Boltzmann theory of conduction [12, 13]. On the other hand, the specific role of long range quasiperiodic order on transport properties is still an open question in spite of a large number of studies (Refs. [8, 14–23] and Refs. therein). In this paper, we study “how electrons propagate” in approximants of the octagonal tiling. This tiling is one of the well-known quasiperiodic tilings that have been used to understand the influence of quasiperiodicity on electronic transport [14–17, 19–21]. The main objective is to show that non standard conduction properties result from purely quantum effects due to quasiperiodicity that cannot be interpreted through the semi-classical theory of transport.

*Corresponding author. Email: guy.trambly@u-cergy.fr

Figure 1. Vertex configurations of the octagonal tiling and of its approximants $k \geq 1$.

2. The octagonal tiling and its approximants

The octagonal, or Ammann-Beenker, tiling [24, 25] is a quasiperiodic tiling analogous to the notorious Penrose tiling. It has eightfold C_{8v} point symmetry, inflation symmetry with ratio $\lambda = 1 + \sqrt{2}$, quasiperiodic translation symmetry, and Bragg peaks indexed by \mathbb{Z}^4 . Those symmetries were observed in quasicrystalline phases of CrNiSi and AlMnSi alloys [26].

A sequence of periodic approximants $X_0, X_1, \dots, X_k, \dots$ can be generated by the inflation mapping M , operating as an invertible integer matrix on \mathbb{Z}^4 [27]. In the physical Euclidean plane E , and in the simplest version where the atoms occupy the tiling vertexes, all the (atomic) sites have integer coordinates in the basis (e_1, e_2, e_3, e_4) , where (e_1, e_2) forms an orthogonal basis of E and (e_3, e_4) is another orthogonal basis related to the former by 45° rotation. All the tiles are congruent to either a square or a rhombus with 45° acute angle.

For $k = 0, 1, 2, \dots$, the k^{th} approximant is periodic, invariant under the translation lattice \mathcal{L}_k generated by $\lambda^k e_1$ and $\lambda^k e_2$ (the square lattice $[e_1, e_2]$ dilated by a factor $L_k = \lambda^k$). In terms of the Pell, or ‘‘octonacci’’, number sequence $P = \{P_k\} = \{0, 1, 2, 5, 12, \dots\}$, defined by $P_0 = 0$, $P_1 = 1$, $P_{k+1} = 2P_k + P_{k-1}$, the unit cell contains $N_k = P_{2k+1} + P_{2k}$ vertexes. Those vertexes are given by the following construction: $\lambda^k(W_k \cap [e_3, e_4])$, *i.e.* select and expand sites of the square lattice $[e_3, e_4]$, eventually bring them back into the square fundamental domain by \mathcal{L}_k translations. The selection window W_k is a semi regular octagon obtained as a projection of the 4D unit cube. Up to a sign $(-1)^k$, W_k is the span (all combinations with coordinates between 0 and 1) of the four vectors

$$P_k(e_4 - e_3), -P_k(e_3 + e_4), (P_k + P_{k-1})e_3, (P_k + P_{k-1})e_4.$$

Ref. [27] provides a slightly more efficient numerical method, avoiding useless scans, by partitioning the octagon into six parallelogram sub-domains; then each selection reduces to specifying ranges for the integer coordinates of a suitable lattice, followed by a global affine transformation.

The first approximant X_0 is a simple square tiling. In all subsequent approximants $k \geq 1$, the configurations around vertexes are the same as in the octagonal quasiperiodic tiling, displayed in Fig. 1. Only the approximants X_1 contain incomplete subsets of these six local patterns, depending on the phase.

In units of edge length $a = \|e_j\|$, the shortest distances between vertexes are $1/\sqrt{2}, 1, \sqrt{2}$, corresponding to the short rhombus diagonal, the tile edge and the square diagonal respectively.

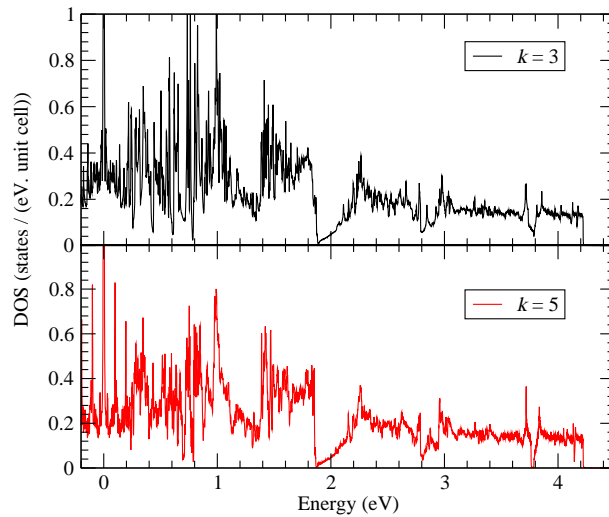


Figure 2. (colour online) Total density of states in perfect octagonal approximants $k = 3$ and $k = 5$. DOS is symmetric w.r.t. $E = 0$ [20].

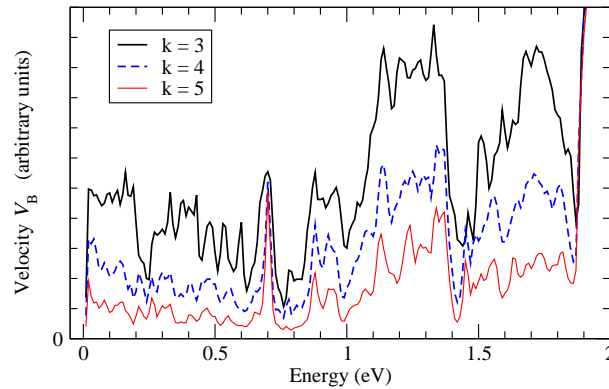


Figure 3. (colour online) Average Boltzmann velocity V_B in perfect octagonal approximants $k = 3$, $k = 4$ and $k = 5$.

3. Electronic structure

We study a pure hopping Hamiltonian

$$H = \gamma \sum_{\langle i,j \rangle} |i\rangle\langle j| \quad (1)$$

where i indexes orbitals located on all vertices and $\gamma = 1$ eV is the strength of the hopping between orbitals. $\langle i, j \rangle$ are the nearest-neighbours at tile edge distance a (figure 1). The properties of this model depend only on topology of the tiling. The total density of states (total DOS), $n(E)$, is computed by a recursion method [22] in a large super-cell containing more than about 10 million orbitals. DOS is shown in figure 2. As already shown by E. S. Zijlstra [20], DOS is spiky at the centre of the band ($|E| < 2$) and smooth near the band edges ($|E| > 2$).

The Boltzmann velocity (intra-band velocity) $V_B \propto \frac{\partial E_n(q)}{\partial q}$ (q : wave vector) is shown figure 3. When the size of the approximant increases, V_B decreases as expected from band scaling analysis [14]. In order to quantify this localisation phe-

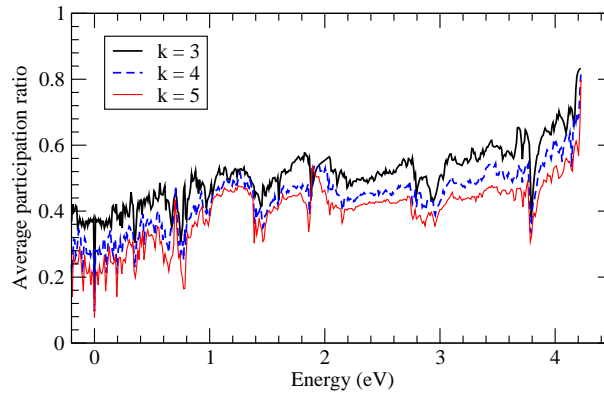


Figure 4. (colour online) Average participation ratio $\bar{p}(E)$ in perfect octagonal approximants $k = 3$, $k = 4$ and $k = 5$.

nomenon, we compute the participation ratio of each eigenstate $|\psi\rangle$ defined by

$$p(\psi) = \left(N \sum_{i=1}^N |\langle i|\psi\rangle|^4 \right)^{-1}, \quad (2)$$

where i indexes orbitals in a \mathcal{L}_k unit cell and $N(\equiv N_k)$ is the number of atoms in this unit cell. For completely delocalised eigenstates p is equal to 1. On the other hand, states localised on one site have a small p value: $p = 1/N$. The average participation ratio $\bar{p}(E)$ of all states with energy close to E are presented on figure 4. This figure shows clearly a stronger localisation of electronic states for larger approximants (larger k values).

4. Quantum diffusion

In the framework of Kubo-Greenwood approach for calculation of the conductivity, the central quantities are the velocity correlation function of states of energy E at time t ,

$$C(E, t) = \left\langle V_x(t)V_x(0) + V_x(0)V_x(t) \right\rangle_E = 2 \operatorname{Re} \left\langle V_x(t)V_x(0) \right\rangle_E, \quad (3)$$

and the average square spreading of states of energy E at time t along the x direction,

$$\Delta X^2(E, t) = \left\langle \left(X(t) - X(0) \right)^2 \right\rangle_E. \quad (4)$$

In (3), $\operatorname{Re} A$ is the real part of A and $V_x(t)$ is the Heisenberg representation of the velocity operator V_x along x direction at time t :

$$V_x = \frac{1}{i\hbar} [X, H]. \quad (5)$$

$C(E, t)$ is related to quantum diffusion by:

$$\frac{d}{dt} \left(\Delta X^2(E, t) \right) = \int_0^t C(E, t') dt'. \quad (6)$$

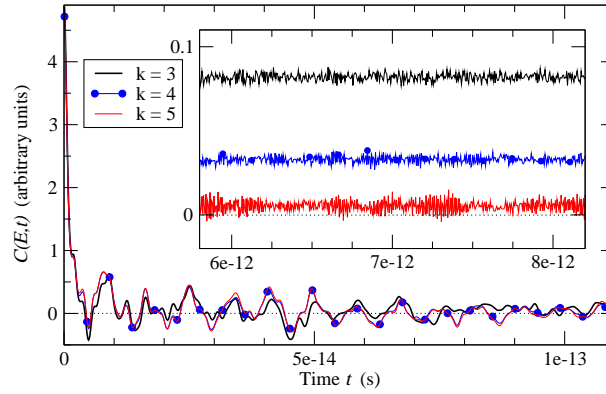


Figure 5. (colour online) Velocity correlation function $C(E, t)$ versus time t in perfect octagonal approximants $k = 3$, $k = 4$ and $k = 5$. For $E = 0.5$ eV.

At zero temperature, the static conductivity is given by the Einstein formula,

$$\sigma = e^2 n(E_F) D(E_F), \quad (7)$$

where E_F is the Fermi energy and D , the diffusivity. In a perfect tiling at temperature $T = 0$ K,

$$D(E) = \frac{\Delta X^2(E, t)}{t} \quad (\text{as } t \rightarrow +\infty). \quad (8)$$

Once the band structure is calculated from the tight-binding Hamiltonian (1) the velocity correlation function can be computed exactly in the basis of Bloch states [12, 28]. Relation (6) shows that an anomalous behaviour of $C(E, t)$ also implies an anomalous behaviour of the quantum diffusion. In crystalline approximants, at the long time, when the spreading of states, $\Delta X = \sqrt{\Delta X^2}$, reaches the size L_k of a unit cell, the propagation should become ballistic. This means that the time averages at long times are $C(E, t) \simeq 2V_B^2$ and $\Delta X^2(E, t) \simeq V_B^2 t^2$, with V_B the Boltzmann velocity along the x direction. But for smaller time values, when $\Delta X < L_k$, the spreading of states depends only on the tiling structure within a unit cell, which is closely related to quasiperiodicity. Therefore, an analysis of electron propagation in approximants on time range before the ballistic regime gives a good indication on how electrons propagate in quasiperiodic structure.

As shown on figure 5, the velocity correlation function in approximants can take negative values at some times, indicating the presence of back-scattering effects in electron propagation. The decay of the Boltzmann term when k increases suggests that, for larger approximants, back-scattering ($C(E, t) < 0$) should be obtained even at some large time values. In that case, conductivity of the tiling will be strongly affected by this back-scattering phenomenon (equation 8).

The average square spreading is the sum of two terms (figure 6):

$$\Delta X^2(E, t) = V_B^2 t^2 + \Delta X_{\text{NB}}^2(E, t). \quad (9)$$

The first term is the ballistic (intra-band) contribution at the energy E . The semi-classical model of Bloch-Boltzmann theory is equivalent to taking into account only this first term. The second term (inter-band contributions) $\Delta X_{\text{NB}}^2(E, t)$ is a non-ballistic (non-Boltzmann) contribution. It is due to the non-diagonal elements of the velocity operator and describes the spreading of the wave-function in a cell. This term is bounded by $L_m(E)^2$, which depends on the energy E [28]. From nu-

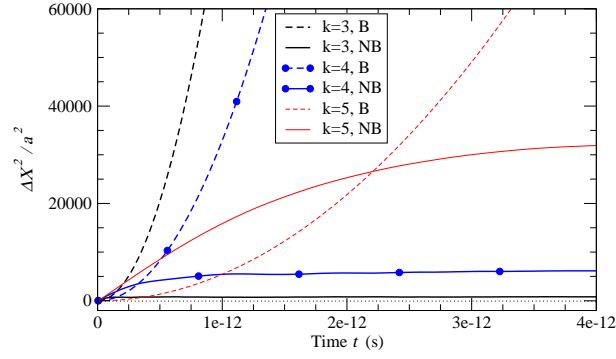


Figure 6. (colour online) Boltzmann (ΔX_B^2) and Non-Boltzmann (ΔX_{NB}^2) square spreading versus time t at $E = 0.5$ eV in perfect octagonal approximants $k = 3$, $k = 4$ and $k = 5$. a is the length of edges of the tiling.

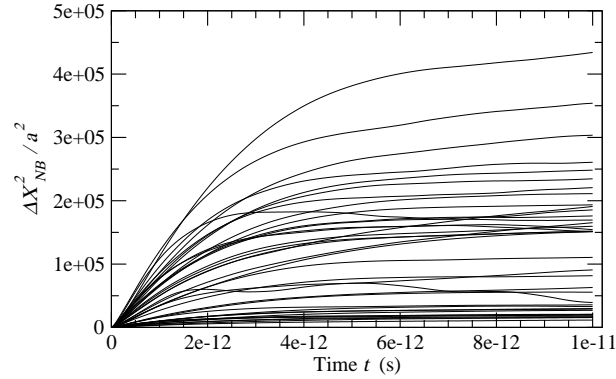


Figure 7. Non-Boltzmann square spreading ΔX_{NB}^2 versus time t at different energies E in perfect octagonal approximant $k = 5$. a is the tile edge length.

merical calculations, it is found that $\Delta X_{NB}^2(E, t)$ reaches rapidly its maximum limit $\sim L_m(E)^2$ (figure 7). Moreover for many energies, numerical calculations show that $L_m(E)$ is of the order of magnitude of the unit cell size L_k or less (see also Ref. [12]). It suggests that in approximants the medium range order localises partially electron states and this phenomenon should lead to anomalous diffusion in quasicrystalline phases. For large time ($t > L_m/V_B$) the ballistic contribution dominates but for small time ($t < L_m/V_B$) the non-ballistic contribution could dominate (“Small velocity regime” [12]). When the size of the approximants increases, the characteristic time L_m/V_B of the crossover between ballistic and non-ballistic behaviour increases.

In the relaxation time approximation [12, 13, 28, 29], the role of phonons and static defects are taken into account by a scattering time τ . τ decreases when temperature T increases and when the number of static defects increases. The velocity correlation function C' at temperature T and/or with static defects is computed from C at $T = 0$ K in perfect tiling,

$$C'(E, t) = C(E, t)e^{-|t|/\tau}. \quad (10)$$

Here the Fermi-Dirac distribution function is taken equal to its zero temperature value. This is valid provided that the electronic properties vary smoothly on the thermal energy scale $k_B T$. The static diffusivity D' is then, [28]

$$D'(E_F, \tau) = \frac{1}{2} \int_0^{+\infty} C(E_F, t)e^{-t/\tau} dt. \quad (11)$$

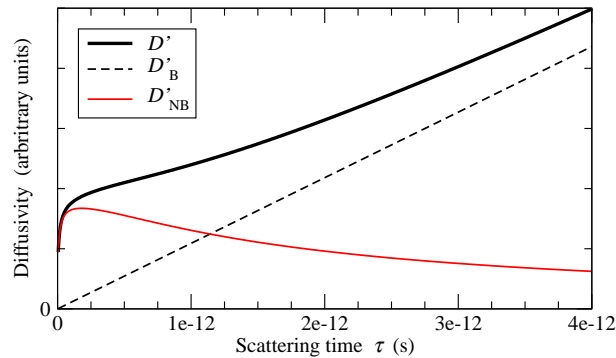


Figure 8. (colour online) Diffusivity D' , in relaxation time approximation, versus scattering time τ , at $E_F = 0.5\text{ eV}$ in octagonal approximant $k = 5$. $D' = D'_B + D'_{NB}$.

Roughly speaking, the transport properties are almost entirely determined by the $\Delta X^2(E, t = \tau)$. Therefore, in large approximants for which ($\tau < L_m/V_B$), transport properties will be governed by non-ballistic (non-metallic) behaviour. Figure 8 shows the diffusivity in approximant $k = 5$, calculated in the relaxation time approximation. D' is the sum of a ballistic term, $D'_B \propto \tau$, and a non-ballistic term, D'_{NB} . For small τ value, its behaviour is not ballistic ($D' \neq D'_B$). In larger approximants ($k > 5$), one can expect an “insulating like” behaviour (D' decreases when τ increases) for a realistic range of τ values, as that found in real approximants [12].

5. Conclusion

To summarise, we have presented quantum diffusion by a pure hopping Hamiltonian in approximants of octagonal tiling ($k \leq 5$). When the size of the approximant increases the usual Boltzmann term decreases rapidly and new non-Boltzmann terms become essential to understand transport properties. These non-Boltzmann terms can have “insulating like” behaviour, suggesting that in larger approximants ($k > 5$), “insulating like” states, due to long range quasiperiodic order, could exist. Calculations in larger approximants are in progress.

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