Sub-diffusive electronic states in octagonal tiling

G. Trambly de Laissardi`ere†, C. Oguey† and D. Mayou∗‡
† Laboratoire de Physique th´eorique et Mod´elisation, CNRS and Université de Cergy-Pontoise, 95302 Cergy-Pontoise, France
∗ Univ. Grenoble Alpes, Inst NEEL, F-38042 Grenoble, France
‡ CNRS, Inst NEEL, F-38042 Grenoble, France
E-mail: guy.trambly@u-cergy.fr

Abstract. We study the quantum diffusion of charge carriers in octagonal tilings. Our numerical results show a power law decay of the wave-packet spreading, \( L(t) \propto t^\beta \), characteristic of critical states in quasicrystals at large time \( t \). For many energies states are sub-diffusive, i.e. \( \beta < 0.5 \), and thus conductivity increases when the amount of defects (static defects and/or temperature) increases.

Experimental investigations have indicated that the conduction properties of many stable quasicrystals (AlCuFe, AlPdMn, AlPdRe, ...) are unusual and differ strongly from those of simple inter-metallic alloys [1-3]. In particular their conductivity increases with static defects density and when temperature increases. It appears also that the medium range order and the chemical order –over one or a few nanometers– have a decisive influence [4-9]. 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 [10-14]. On the other hand, the specific role of long range quasiperiodic order in electronic properties is still an open question in spite of a large number of studies (Refs. [15-37] and Refs. therein). Many studies support the existence of critical states, which are neither extended nor localised, but are characterised by a power law decay of the wave-function envelope at large distances. In the presence of critical states, the diffusion of charge carrier at sufficiently large time \( t \) follows a power law and then the spatial extension \( L \) of wave-packets should be, \[ L(E, t) \propto t^{\beta(E)} \text{ at large } t, \] where \( \beta, 0 \leq \beta \leq 1 \), is an exponent depending on energy \( E \) and on the Hamiltonian model. Note that in usual metallic crystals without static defects, \( \beta = 1 \) and the propagation is ballistic. In strongly disordered systems, \( \beta = 0.5 \) for a large time range and propagation is diffusive. For a localised state one has \( \beta = 0 \). When disorder is introduced in the perfect approximant or perfect quasicrystal in the form of static defects (elastic scatterers) and/or inelastic scattering (temperature, magnetic field...), the defects induce scattering and we expect that there is an associated time \( \tau \) above...
which the propagation of the wave-packet is diffusive. The diffusivity \( D \) of charge carrier at energy \( E \) can be estimated by, 
\[
D(E, \tau) \simeq L(E, t = \tau)^2 / \tau \propto \tau^{2\beta(E) - 1},
\]
and the conductivity \( \sigma \) at zero frequency is given by the Einstein formula:
\[
\sigma(E_F, \tau) = e^2 n(E_F) D(E_F, \tau) \propto \tau^{2\beta(E_F) - 1},
\]
where \( n \) is the density of states and \( E_F \) the Fermi energy. The case \( 0.5 < \beta < 1 \), called super-diffusive regime, leads to transport properties similar to metal, since the conductivity decreases when disorder increases – i.e. when \( \tau \) decreases –. Conversely, for \( 0 < \beta < 0.5 \), the regime is sub-diffusive and the conductivity increases when disorder increases like in real quasicrystals. Many authors consider [18-20, 29, 34-37] that critical states could lead to \( \beta < 0.5 \) but it has not yet been shown in 2D or 3D quasiperiodic structures (except for some very specific energies).

**Model Hamiltonian.**– The octagonal, or Ammann-Beenker, tiling [38, 39] is a quasiperiodic tiling analogous to the notorious Penrose tiling. This tiling has been often used to understand the influence of quasiperiodicity on electronic transport [18-25]. A sequence of periodic approximants \( X_0, X_1, \ldots, X_k, \ldots \) can be generated [40]. In approximants of order \( k \geq 1 \), the 6 local configurations around vertexes are the same as in the octagonal quasiperiodic tiling. They have, respectively, coordination number \( \eta = 3, 4, 5, 6, 7 \) and 8. We consider the simple Hamiltonian,
\[
\hat{H} = \sum_i \epsilon_i c_i^* c_i + \sum_{\langle ij \rangle} \gamma c_i^* c_j + h.c.,
\]
where \( i \) indexes \( s \) orbitals located on vertexes, and \( \gamma \) is the strength of the hopping between orbitals. \( \langle i, j \rangle \) are the nearest-neighbours at tile edge distance \( a \). To simulate schematically a possible effect of the presence of different chemical elements, the one-site energy \( \epsilon_i \) is proportional to the coordinance \( \eta_i \) of the site \( i \): \( \epsilon_i = \eta_i \gamma \). To obtain realistic time values, we use \( \gamma = 1 \) eV which is the order of magnitude of the hopping parameter in real inter-metallic compounds. The total density of states (total DOS) of \( X_7 \) approximant is shown figure 1(a).

**Quantum diffusion.**– In the framework of Kubo-Greenwood approach for calculation of the conductivity, we use the polynomial expansion method developed by Mayou, Khenna, Roche and Triozon [41, 42, 31, 27, 28] to compute the mean square spreading of the wave-packet at time \( t \) and energy \( E \): 
\[
L^2(E, t) = \langle (\hat{X}(t) - \hat{X}(0))^2 \rangle_E,
\]
where \( \hat{X} \) is the position operator in the \( x \)-direction. The diffusion coefficient \( \mathcal{D}(E, t) = L(E, t)^2 / t \) is shown in figure 1(b) for \( X_7 \) approximant at some energies. The ballistic regime due to the periodicity of the approximant is reached at very large \( t \), when \( L(t) > L_k \) where \( L_k \) is the approximant cell size; then \( L(t) = V_B t \), where \( V_B \) is the Boltzmann velocity, i.e. the intra-band velocity, \( V_B = \langle \partial E_n(k) / \partial k_x \rangle_E / \hbar \), where \( E_n(k) \) is the band dispersion relation [12]. For \( X_7 \) in time range shown figure 1, this Boltzmann term is negligible and, for all purposes of this discussion, the \( X_7 \) approximant is equivalent to the quasiperiodic system. Depending on the \( t \) values, three different regimes are observed at each energy:

- At very small time, typically when \( L(t) < a \), the mean spreading grows linearly with \( t \), \( L(t) = V_0 t \) (ballistic behaviour), where \( V_0 > V_B \) [10, 12].
- For times, corresponding to \( L(t) \gtrsim a \), the propagation seems to become diffusive as the diffusion coefficient is almost constant, \( \mathcal{D}(t) \simeq \mathcal{D}_{diff} \). Therefore \( L_1 \),
defined by $L_1 = D_{\text{diff}}/V_0 \simeq$ a few $a$, is a kind of effective elastic scattering length but it is not due to static scattering events because we consider perfect tilings. The corresponding effective elastic scattering time is $t_1 = L_1/V_0$. Roughly speaking, it seems that when $L(t) \gtrsim L_1$, i.e. $t \gtrsim t_1$, the wave-packet feels a random tiling.

- An other distance $L_2$ (respectively an other time $t_2$, $L(t_2) = L_2$) appears. For $L(t) > L_2 \simeq$ a few $10a$, a new regime appears and $D(t)$ follows a power law. It is thus characteristic of the medium and long range quasiperiodic order. Figure 1(b) shows that the $\beta$ value can switch from a sub-diffusive regime ($\beta < 0.5$) to a super-diffusive regime ($\beta > 0.5$) over a small variation of energy. The $t_2$ values, $t_2 \simeq 10^{-13} - 10^{-14}$ s, have the order of magnitude of the scattering time above which measurements show unusual transport properties in quasicrystals [1].

Both distances $L_1 \simeq$ a few $a$, $L_2 \simeq$ a few $10a$, and the exponent $\beta$ at time $t > t_2$, depend a lot on the energy value $E$. $L_1 < L_2$, but at some energy it even seems that $L_1 \simeq L_2$. Further analysis are necessary to understand the energy dependence.

To summarise, we have presented quantum diffusion in a large approximant of the octagonal tiling. The charge carrier propagation is determined by the wave-packet spreading in the quasiperiodic lattice. From numerical calculation, two length scales seem to characterise this quasiperiodic spreading. $L_1$, typically $L_1 = a$ few $a$, above which the propagation is almost diffusive in spite of the absence of static defects. $L_2$, typically $L_2 = a$ few $10a$, above which specific quasiperiodic symmetries lead to a power law dependence of the root mean square spreading, $L(t) \propto t^\beta$. For some energies states are super-diffusive or diffusive, i.e. $\beta \geq 0.5$, whereas for other energies, a sub-diffusive regime, i.e. $\beta < 0.5$, sets in as expected for critical states characteristic of quasiperiodicity. This sub-diffusive regime is the generalisation to quasicrystal of the
non-Boltzmann propagation found in realistic approximants of i-AlMnSi and i-AlCuFe [10, 11], in the complex inter-metallic alloys λ-AlMn [12], and in small approximants of octagonal and Penrose tilings [13, 14].

The computations were performed at the Centre de Calcul (CDC), Université de Cergy-Pontoise. We thank Y. Costes for computing assistance.

References

[1] Berger C 1994 Lectures on Quasicrystals, ed F Hippiert and D Gratias (Les Ulis, Les Editions de Physique) p 463
[2] Grenet T 2000 Quasicrystals: Current Topics, ed E Belin-Ferré, C Berger, M Quiquandon and A Sadoc (Singapore 2000, World Scientific) p 455
[3] Delahaye J and Berger C 2015 Eur. Phys. J. B 88 102
[4] Fujiwara T, Yamamoto S and Trambly de Laisserdère G 1993 Phys. Rev. Lett. 71 4166
[5] Krajčí M et al. 1995 Phys. Rev. B 51 17355
[6] Fujiwara T, Mitsui T and Yamamoto S 1996 Phys. Rev. B 53 R2910
[7] Roche S and Fujiwara T 1998 Phys. Rev. B 58 11338
[8] Trambly de Laisserdère G and Mayou D 1998 Phys. Rev. B 55 9157
[9] Roche S and Fujiwara T 1999 Phys. Rev. B 60 322
[10] Fujiwara T, Kohmoto M and Tokihiro T 1989 Phys. Rev. B 40 7413
[11] Sire C and Bellissard J 1990 Europhys. Lett. 11 439
[12] Trambly de Laisserdère G and Mayou D 1995 Phys. Rev. B 51 15827
[13] Roche S and Mayou D 1997 Phys. Rev. Lett. 79 2518
[14] Sire C 1994 Lectures on Quasicrystals, ed F Hippiert and D Gratias (Les Ulis, Les Editions de Physique) p 505