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Numerical evidence of backscattering in approximants of quasicrystals

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We compute the velocity correlation function of electronic states close to the Fermi energy, in approximants of quasicrystals. As we show the long time value of this correlation function is small. This means a small Fermi velocity, in agreement with previous band structure studies. Furthermore the correlation function is negative on a large time interval which means a phenomenon of backscattering. As shown in previous studies the backscattering can explain unusual conduction properties, observed in these alloys, such as for example the increase of conductivity with disorder.

Keywords: quasicrystals; transport properties; quantum diffusion

1 Introduction

Quasicrystals have many fascinating electronic properties, and in particular quasicrystals with high structural quality, such as the icosahedral AlCuFe and AlPdMn alloys, have unconventional conduction properties when compared with standard intermetallic alloys. Their conductivities can be as low as $150\text{--}200 (\Omega \text{ cm})^{-1}$. Furthermore the conductivity increases with disorder and with temperature, a behaviour just at the opposite of that of standard metal. In a sense the most striking property is the so-called “*inverse Mathiessen rule*” according to which the *increases of conductivity* due to different sources of disorder seems to be *additive*. This is just the opposite that happens with normal metals where the increases of resistivity due to several sources of scattering are *additive*. Finally the Drude peak which is a signature of a normal metal is also absent in the optical conductivity of these quasicrystals.

An important result is also that many approximants of these quasicrystalline phases have similar conduction properties. For example the crystalline α -AlMnSi phase with a unit cell size of about 12 \AA and 138 atoms in the unit cell has a conductivity of about $300 (\Omega \text{ cm})^{-1}$ at low temperature [1, 2]. The conductivity has the same defect and temperature dependence as that of the AlCuFe and AlPdMn icosahedral phase. There is, to our knowledge, no experimental result on the optical conductivity of this α -AlMnSi phase, but it is very likely that it is similar to that of AlCuFe and AlPdMn icosahedral phase.

The interpretation of these unconventional conduction properties is still a challenge for condensed matter physicists. Several models have been proposed including thermally activated hopping [3], band structure effects due to small density of states and narrow

pseudo-gap [4, 5] of anomalous quantum diffusion [6, 7, 8, 9, 10, 11, 12]. Yet all these models are difficult to compare in a quantitative way with experiments. In this paper we present preliminary results of an *ab-initio* study of transport in the crystalline α -AlMnSi phase. The number of atoms in the unit cell (138) is sufficiently small to permit computation with the *ab-initio* L.M.T.O. method and provides us a good starting model. Obviously, this approach has still limitations due to the local density approximation (L.D.A.) treatment of electron correlations and due to the approximation in the solution of the Schrödinger equation. Yet we believe that this starting point is much better than simplified s-bands model of tight-binding type.

In this paper we consider the quantum diffusion properties of the perfect α -AlMnSi crystal and show that they are quite unusual. The central quantities are the velocity correlation function of states of energy E at time t : $C(E, t)$, and the average square spreading of states of energy E at time t along the x direction: $X^2(E, t)$. The velocity correlation function is defined by:

$$C(E, t) = \left\langle V_x(t)V_x(0) + V_x(0)V_x(t) \right\rangle_E = 2 \operatorname{Re} \left\langle V_x(t)V_x(0) \right\rangle_E, \quad (1)$$

where $\langle A \rangle_E$ is the average of the operator A on states of energy E . $\operatorname{Re} B$ is the real part of B and $V_x(t)$ is the Heisenberg representation of the velocity operator along x direction at time t . $C(E, t)$ is related to quantum diffusion by:

$$\frac{dX^2(E, t)}{dt} = \int_0^t C(E, t') dt'. \quad (2)$$

Once the bandstructure is calculated in a self-consistent way, the velocity correlation function can be computed exactly in the basis of Bloch states. Relation (2) shows that an anomalous behaviour of $C(E, t)$ implies also an anomalous behaviour of the quantum diffusion which is the basis for the model [10, 12] of conductivity of quasicrystals. In the long time limit one knows that the propagation is ballistic, this means that $X^2(E, t)$ is given by $v_F^2 t^2$ at large time. From (2) one deduces that $C(E, t)$ is of the order of $2v_F^2$ at large time. But at intermediate times (see below) the behaviour of $C(E, t)$ is more specific of the crystal and we show that the α -AlMnSi phase is different compared to other good metals such as Al (f.c.c.), cubic Al_{12}Mn ...

In particular we find that there is on the average a phenomenon of *backscattering* in α -AlMnSi phase. This means that the velocity correlation function is often negative. This negative value has been shown previously [12] a sufficient condition to explain the unusual conduction properties of these alloys.

2 *Ab-initio* electronic structure

Electronic structure determinations have been performed in the frame-work of density functional formalism in the local density approximation (L.D.A.) by using the self-consistent Tight-Binding (T.B.) Linear Muffin Tin Orbital (L.M.T.O.) method in the Atomic Sphere Approximation (A.S.A.) [13]. The LMTO basis includes all angular moments up to $l = 2$ and the valence states are Al (3s, 3p, 3d), Mn (4s, 4p, 3d).

The LMTO DOS of an α -AlMn idealized approximant (Elser-Henley model [14]) has been first calculated by T. Fujiwara [15, 5]. This original work shows the presence of a Hume-Rothery pseudo-gap near the Fermi energy, E_F , in agreement with experimental

results [4, 2]. The role of the transition metal (TM) elements in the pseudo-gap formation has also been shown from *ab-initio* calculations [16] and experiments. Indeed the formation of the pseudo-gap results from a strong sp-d coupling associated to an ordered sub-lattice of TM atoms. Just as for Hume-Rothery phases a description of the band energy can be made in terms of pair interactions. We have shown that a medium-range Mn-Mn interaction mediated by the sp(Al)-d(Mn) hybridization plays a determinant role in the occurrence of the pseudo-gap [17, 16]. It is thus essential to take into account the chemical nature of the elements to analyze the electronic properties of approximants. E.S. Zijlstra and S.K. Bose [18] performed a detailed *ab-initio* study of the role of Si atoms substitution and vacancies in the electronic structure of α -AlMn. They show that Si atoms are in substitution with some Al atoms. The main effect of Si is to shift E_F in the pseudo-gap in agreement with Hume-Rothery mechanism to minimize band energy.

The electronic structures of simpler crystals such as orthorhombic Al_6Mn , cubic Al_{12}Mn , present [16] also a pseudo-gap near E_F but it is less pronounced than for complex approximant phases.

3 Velocity correlation function

Starting from the self-consistent LMTO eigenstate Ψ_n with energy E_n , the velocity correlation function is [10]:

$$C(E, t) = 2 \left\langle \text{Re} \left\{ e^{i \frac{E_n t}{\hbar}} \langle \Psi_n | V_x e^{-i \frac{H t}{\hbar}} V_x | \Psi_n \rangle \right\} \right\rangle_{E_n=E} \quad (3)$$

By using the closure properties of the eigenstates one obtains easily:

$$C(E, t) = 2 \left\langle \sum_p \cos \left((E_n - E_p) \frac{t}{\hbar} \right) \left| \langle \Psi_n | V_x | \Psi_p \rangle \right|^2 \right\rangle_{E_n=E} \quad (4)$$

where the sum is over all the eigenstates Ψ_p with the same vector \vec{k} than Ψ_n . In (4) the terms $n = p$ are the Boltzmann contribution to the velocity correlation function:

$$C_B(E, t) = 2 \left\langle \sum_n \left| \langle \Psi_n | V_x | \Psi_n \rangle \right|^2 \right\rangle_{E_n=E} = 2v_B^2 \quad (5)$$

which does not depend on the time t . The products $\langle \Psi_n | V_x | \Psi_p \rangle$ are calculated from L.M.T.O. eigenstates by using a numerical derivation of the hamiltonian in the reciprocal space. We compute $C(E, t)$ for crystals (complex approximants and simple crystals), where eigenstates are Bloch states depending on vector \vec{k} in the first Brillouin zone. In equations (4)–(5), the average on states of energy E is obtained by taking the eigenstates of each \vec{k} vector with an energy E_n such as: $E - \frac{1}{2}\Delta E < E_n < E + \frac{1}{2}\Delta E$. ΔE is a kind of energy resolution of the calculation. The calculated $C(E, t)$ is rather sensible to small number N_k of \vec{k} vectors in the first Brillouin zone. Therefore N_k increases until $C(E, t)$ does not depend significantly on N_k .

$C(E_F, t)$ for Al (f.c.c), cubic Al_{12}Mn [19], and the cubic approximant α - $\text{Al}_{114}\text{Mn}_{24}$ (with the experimental atomic structure [20]) are shown in figure 1. E_F is the Fermi energy calculated by the self-consistent L.M.T.O. procedure. ΔE is fixed arbitrarily to $\Delta E = 1.36$ eV. The number of \vec{k} points in the first Brillouin zone is $N_k = 80^3$, 40^3 and 10^3 for

For large t ,

$$\lim_{t \rightarrow +\infty} C(E_F, t) \simeq C_B(E_F, t) = 2v_F^2, \quad (6)$$

where v_F is the Boltzmann velocity (intra-band velocity) at the Fermi energy: $v_F = 9.6 \cdot 10^7$, $3.4 \cdot 10^7$, and $6.2 \cdot 10^6$ cm s⁻¹, for Al, Al₁₂Mn and α -Al₁₁₄Mn₂₄, respectively. This last result is very similar to the original work of T. Fujiwara et al. [5] for the α -Al₁₁₄Mn₂₄ (with the atomic structure model of Elser-Henley). The strong reduction of v_F in the approximant phase with respect to simple crystal phases shows the importance of the medium-range order (up to distances equal to 12–20 Å). This leads to a very small Boltzmann conductivity for the approximant [5]. Since the medium-range order in approximant is a consequence of the quasiperiodic order [22], these results suggest that the backscattering is a consequence of quasiperiodic order.

When t is finite (figure 1), $C(E_F, t)$ and $C_B(E_F, t)$ differ, and there is a new difference between approximant and simple crystal. In the case of Al (f.c.c.) phase, $C(E_F, t)$ is always positive, and the Boltzmann value is reached rapidly when t increases. But for some t values the velocity correlation function $C(E_F, t)$ is negative for Al₁₂Mn and α -Al₁₁₄Mn₂₄. That means that at these times the phenomenon of *backscattering* occurs.

Roughly speaking, the transports properties depends on the values of $C(E_F, t)$ over all times t from 0 to the scattering time τ [10, 12] (see for instance equation (2)). A realistic value of τ has been estimated to about 10^{-14} s [21]. For the simple crystals Al₁₂Mn, $C(E_F, t)$ is meanly positive when $t > 2 \cdot 10^{-15}$ s. But for the complex approximant α -Al₁₁₄Mn₂₄, a lot of t values correspond to $C(E_F, t) < 0$, even when t is close to τ or larger (figure 2). Therefore, in the case of Al₁₂Mn, the backscattering (negative range of $C(E_F, t)$) should have a negligible effect on the transport properties, whereas this effect must be determinant for the approximant.

4 Conclusion

We present *ab-initio* calculations of the velocity correlation function of the electronic states close to the Fermi energy, in a complex approximant and simple crystals. These calculations are the first numerical proof of the existence of the phenomenon of backscattering in an approximant of quasicrystals. This shows that a Boltzmann approach is not enough to understand the unusual transport properties of quasicrystals. Calculations of the electrical conductivity are in progress.

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Figure 1: Velocity correlation function $C(E_F, t)$ ($\text{m}^2 \text{s}^{-2}$) versus time t (s), for (a) Al (f.c.c), (b) cubic Al_{12}Mn , and (c) cubic approximant $\alpha\text{-Al}_{114}\text{Mn}_{24}$. The dashed lines are the Boltzmann velocity correlation function $C_B(E_F, t) = 2v_F^2$.

Figure 2: Velocity correlation function $C(E_F, t)$ ($\text{m}^2 \text{s}^{-2}$) versus large time t (see figure 1). The dashed lines are the corresponding Boltzmann velocity correlation function $C_B(E_F, t) = 2v_F^2$. The little dots line is the x-axis ($C(E_F, t) = 0$).

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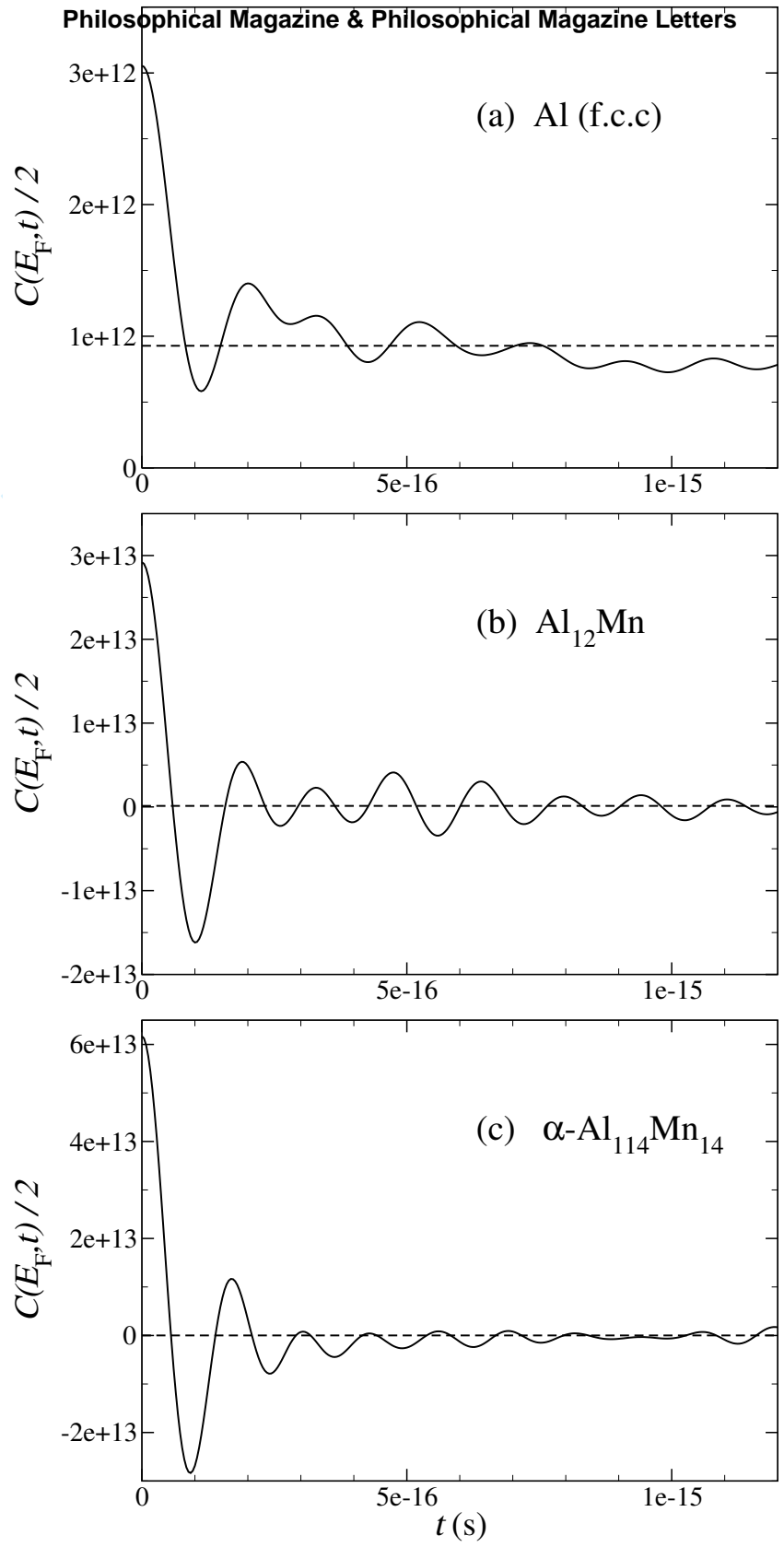


figure 1.

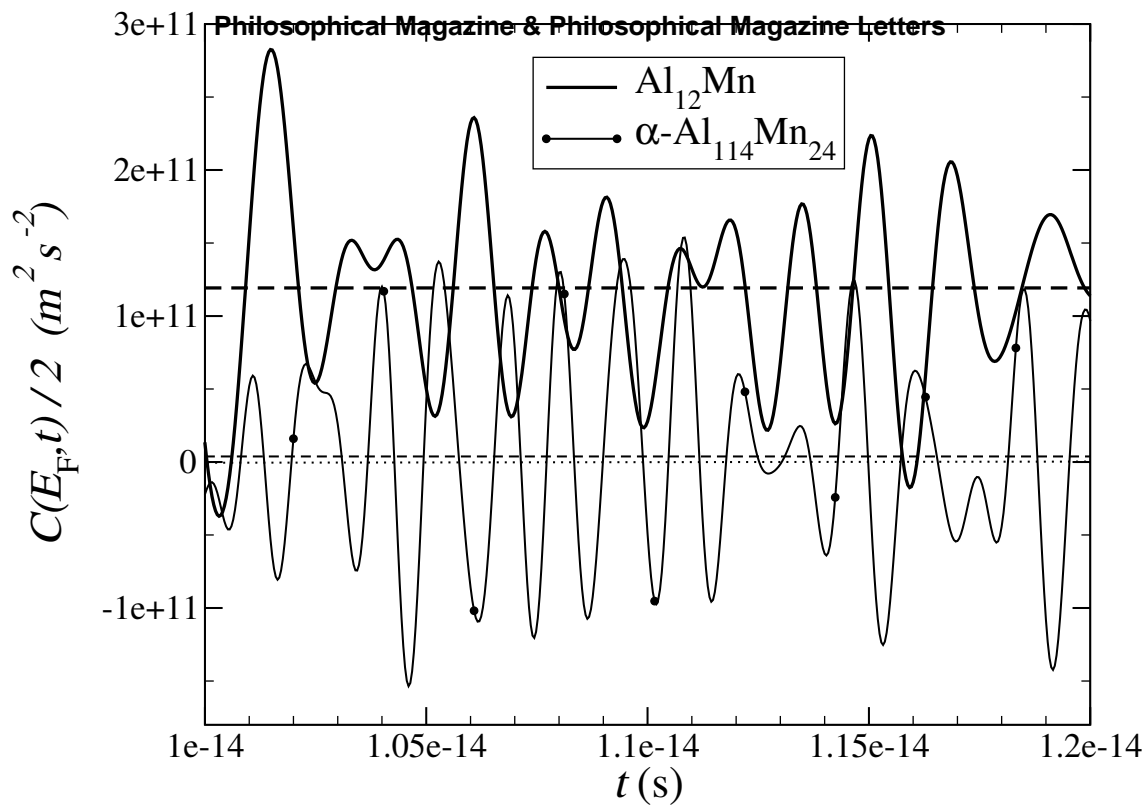


figure 2.