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Quantum transport of slow charge carriers in quasicrystals and correlated systems

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We show that the semi-classical model of conduction breaks down if the mean free path of charge carriers is smaller than a typical extension of their wavefunction. This situation is realized for sufficiently slow charge carriers and leads to a transition from a metallic like to an insulating like regime when scattering by defects increases. This explains the unconventional conduction properties of quasicrystals and related alloys. The conduction properties of some heavy fermions or polaronic systems, where charge carriers are also slow, present a deep analogy.

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The semi-classical Bloch-Boltzmann theory, which plays a fundamental role in our understanding of conduction in solids, has limitations such as for example magnetic and electric breakdown [1] or quantum interferences in the diffusive regime [2] that are well known and have been intensively studied. The present work focuses on another limitation that has received little attention and that occurs when charge carriers have sufficiently small velocities.

Indeed the semi-classical theory of conduction in crystals is based on the concept of a charge carrier wave-packet propagating at a velocity $V = (1/\hbar)\partial E_n(k)/\partial k$, where “ $E_n(k)$ ” is the dispersion relation for band n and wavevector k . The validity of the wave-packet concept requires that the extension L_{wp} of the wave-packet of the charge carrier is smaller than the distance $V\tau$ of traveling between two scattering events separated by a time τ . On the contrary, if $V\tau < L_{wp}$, a condition that can be realized by sufficiently slow charge carriers, the semi-classical model breaks down. Here we report on a quantum theory that allows to treat on the same footing the standard regime where the semi-classical approach is valid and the regime of slow charge carriers. We find that when the scattering time τ decreases a transition can occur between a metallic regime for $V\tau > L_{wp}$ and an insulating like regime for $V\tau < L_{wp}$. As an example we consider specifically a complex metallic alloy: the α -AlMnSi phase. Ab-initio band structure calculations show that the samples of this system, that have been studied experimentally, are in the small velocity regime $V\tau < L_{wp}$. This explains their unconventional conduction properties. The α -AlMnSi phase is structurally related to the icosahedral quasicrystalline phase AlMnSi and shares many similar conduction properties with other icosahedral phases such as AlCuFe and AlPdMn and their crystalline approximants [3, 4, 5]. Thus the present work is relevant for these systems too and gives a strong insight in the so far unexplained properties of this class of materials. The concepts developed here open also a

new insight in the physics of correlated systems. Indeed recent studies of some heavy fermions or polaronic systems [6, 7, 8], where charge carriers are also slow, show that their conduction properties present a deep analogy with those described here. In particular a transition is observed from a metallic like regime at low temperature (weak scattering) to an insulating like regime at higher temperature (stronger scattering).

We come now to the treatment of the conductivity of independent electrons. According to the Einstein relation the conductivity σ depends on the diffusivity $D(E)$ of electrons of energy E and the density of states $n(E)$ (summing the spin up and spin down contribution). We assume that $n(E)$ and $D(E)$ vary weakly on the thermal energy scale kT , which is justified here. In that case, the Einstein formula writes $\sigma = e^2 n(E_F) D(E_F)$, where E_F is the chemical potential and e is the electronic charge. The temperature dependence of σ is due to the variation of the diffusivity $D(E_F)$ with temperature. The central quantity is thus the diffusivity which is related to quantum diffusion.

For a time independent hamiltonian, the quantum diffusion of states at energy E can be measured through the square of their spreading defined as :

$$\Delta X^2(E, t) = \left\langle (X(t) - X(0))^2 \right\rangle_E \quad (1)$$

$X(t)$ is the Heisenberg representation of the position of the electron along a chosen axis “ x ”. $\langle \rangle_E$ means an average over all states with energy equal to E . The diffusivity is given by

$$D(E) = \frac{1}{2} \lim_{t \rightarrow \infty} \frac{d\Delta X^2(E, t)}{dt}. \quad (2)$$

$\Delta X^2(E, t)$ can be computed through the velocity correlation $C(E, t) = \left\langle V_x(t)V_x(0) + V_x(0)V_x(t) \right\rangle_E$. $C(E, t)$ measures the correlation between the velocities at time t_0 and $t_0 + t$ and is independent of t_0 . It can be shown

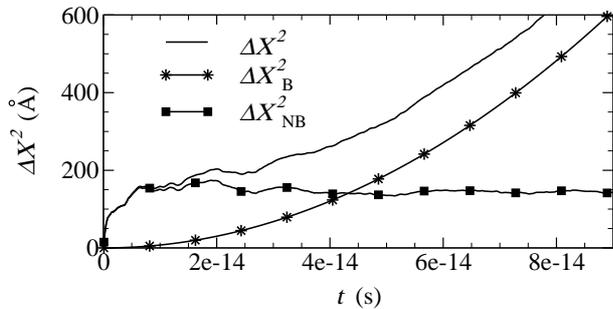


FIG. 1: Square spreading ΔX^2 of electrons states with Fermi energy E_F versus time t , in the cubic approximant α -Al_{69.6}Si_{13.0}Mn_{17.4}. ΔX^2 is the sum of a Boltzmann term, $\Delta X_B^2 = V_F^2 t^2$, and a non Boltzmann term ΔX_{NB}^2 .

that [9] :

$$\frac{d\Delta X^2(E, t)}{dt} = \int_0^t C(E, t') dt', \quad (3)$$

which allows to compute $\Delta X^2(E, t)$ in terms of the velocity correlation function $C(E, t)$ with the initial condition $\Delta X^2(E, t = 0) = 0$. For a perfect crystal $C(E, t)$ is given by $C(E, t) = \langle C(n\vec{k}, t) \rangle_E$ where $|n\vec{k}\rangle$ are Bloch states with energy $E_n(\vec{k}) = E$ and:

$$C(n\vec{k}, t) = 2 \sum_m |V_{n,m}(\vec{k})|^2 \cos\left(\frac{(E_n(\vec{k}) - E_m(\vec{k}))t}{\hbar}\right). \quad (4)$$

$V_{n,m}(\vec{k})$ are matrix elements of the velocity operator V_x between $|n\vec{k}\rangle$ and $|m\vec{k}\rangle$. In the sum (4), the diagonal elements ($m = n$) are time independent, whereas the other terms depend on t . In the perfect crystal $\Delta X^2(E, t)$, which is linearly related to $C(E, t)$ by (3), is the sum of two terms:

$$\Delta X^2(E, t) = V^2 t^2 + \Delta X_{NB}^2(E, t). \quad (5)$$

The first term is the ballistic contribution where “ V^2 ” is the average of $|V_{n,n}(\vec{k})|^2$ over all states $|n\vec{k}\rangle$ at the energy E . The semi-classical theory is equivalent to take into account only this first term. The second term in (5), $\Delta X_{NB}^2(E, t)$, is a non ballistic (non Boltzmann) contribution. It is due to the non diagonal elements of the velocity operator ($m \neq n$, in (4)) and it describes the spreading of the wavefunction in the unit cell. It can be shown that $\Delta X_{NB}^2(E, t) < L^2$, where L is the unit cell length along the x axis. In presence of a disorder characterized by a scattering time τ such that the non ballistic term dominates i.e. $\Delta X_{NB}^2(E, \tau) > V^2 \tau^2$, the semi-classical theory breaks down and the conductor is a non standard metal.

An example of such a crossover from a standard (ballistic) behavior to a non standard (non ballistic) behavior is shown on Fig. 1 in the case of the Fermi energy states of the complex phase α -AlMnSi (see below for more details). The non Boltzmann term $\Delta X_{NB}^2(t)$ increases very

rapidly and saturates to a maximum value of the order of the square size of the unit cell. At small time t , the standard Boltzmann contribution, $\Delta X_B^2(t) = V^2 t^2$, is smaller due to a very small velocity V of the electrons. Thus α -AlMnSi is a non conventional metal at these time scale i.e. when the scattering time τ is less than $\sim 3-4 \cdot 10^{-14}$ s. By contrast in pure Al, ΔX_B^2 is typically 10^3 times larger and ΔX_{NB}^2 is about 10 times smaller. Therefore the non Boltzmann contribution is negligible at these time scales and Al is a standard metal.

For a system with disorder (static disorder and/or temperature dependent scattering), we compute the diffusivity within a relaxation time approximation [9] :

$$C'(E, t) = C(E, t) \exp(-t/\tau), \quad (6)$$

where C is the velocity correlation function of the perfect crystal and C' is that of the system with defects. A relaxation time τ is defined, which meaning is the following. After (6) and (3), at a time scale $t < \tau$ the propagation of wavefunctions is not affected by disorder and is that of the perfect crystal, but at a time scale $t > \tau$ the propagation becomes diffusive due to the disorder. (3), (6) and (2) lead to the expressions of the diffusivity D and of the dc-conductivity σ of the system with defects:

$$D = V^2 \tau + \frac{L^2(\tau)}{\tau}, \quad (7)$$

$$\sigma = e^2 n(E_F) V^2 \tau + e^2 n(E_F) \frac{L^2(\tau)}{\tau}. \quad (8)$$

Thus D and σ have two parts. The first ones, named in the following D_B and σ_B , are the usual Bloch-Boltzmann results, and the second ones, named D_{NB} and σ_{NB} , are due to the non Boltzmann contributions. $L^2(\tau)$ is a proper average of $\Delta X_{NB}^2(E_F, t)$ on a time scale τ . The second term in the right hand side of (8) is equivalent to the conductivity of Anderson insulators in the Thouless regime [10, 11]. Indeed in this regime the wavefunction spreads between two inelastic scattering events but the spreading saturates to the localization length. Here the localization length is replaced by the large time limit of $L(t)$ which, as can be shown, is bounded by the unit cell length L .

We come now to a detailed analysis of the unusual transport properties of the cubic 1/1 icosahedral approximant AlMnSi [12]. These results will be compared with those of pure Al (f.c.c.), cubic Al₁₂Mn [13] and orthorhombic Al₆Mn [13] crystals, which have a metallic behavior. For all those systems, the electronic structure calculation have been performed [13, 14, 15] by using the self-consistent Tight-Binding Linear Muffin Tin orbital (TB-LMTO) method [16]. Starting from the computed eigenstates $|n\vec{k}\rangle$, we use equations (3) and (4) to compute the velocity correlation function without disorder, $C(E, t)$ and the square of the spreading $\Delta X^2(E, t)$. The average $\langle \rangle_E$ is obtained by taking the eigenstates of each \vec{k} vector with an energy $E_n(\vec{k})$ such as: $E - \Delta E/2 < E_n(\vec{k}) < E + \Delta E/2$. ΔE is an energy resolution in

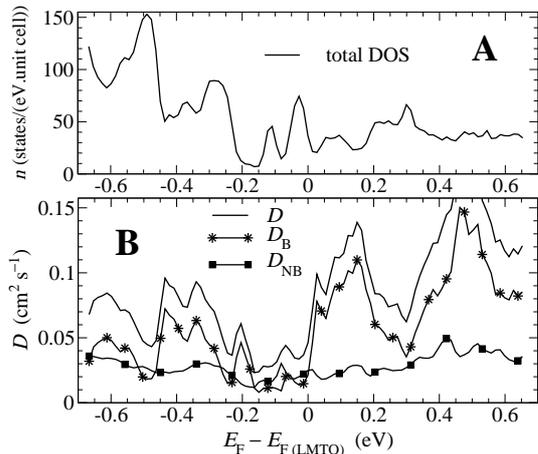


FIG. 2: (A) LMTO total DOS $n(E_F)$ and (B) diffusivity $D(E_F)$ in the cubic approximant $\alpha\text{-Al}_{69.6}\text{Si}_{13.0}\text{Mn}_{17.4}$ ($\tau = 3 \cdot 10^{-14}$ s). D is the sum of a Boltzmann term, D_B , and a non Boltzmann term, D_{NB} (see equation (7)).

the calculation. The number of k-points per first Brillouin zone included in this average calculation is N_k . For a given ΔE , N_k is increased until C does not depend significantly on N_k . Therefore, there is not adjustable parameter in our calculation. For Al (f.c.c.), Al_{12}Mn , Al_6Mn and $\alpha\text{-Al}_{69.6}\text{Si}_{13.0}\text{Mn}_{17.4}$, ΔE is equal to 0.272; 0.272; 0.272; 0.0272 eV, respectively, and N_k is equal to 1,728,000; 64,000; 18,000; 32,768; respectively.

For the $\alpha\text{-AlMnSi}$ phase, we use the experimental atomic structure [12] and the Si positions proposed by E. S. Zijlstra and S. K. Bose [15] to calculate transport properties in an α -phase with composition: $\alpha\text{-Al}_{69.6}\text{Si}_{13.0}\text{Mn}_{17.4}$. This phase contains 138 atoms in a cubic unit cell: 96 Al atoms, 18 Si atoms, and 24 Mn atoms. In Fig. 2, the total DOS n of the $\alpha\text{-AlMnSi}$ phase is presented versus the energy. Its total density of states is characterized [14] by a depletion near the Fermi energy E_F , called pseudo-gap, which is observed experimentally. This is due to a Hume-Rothery mechanism [13, 17] of band energy minimization that is strong in quasicrystals and their approximants [17]. Following the Hume-Rothery condition, it is expected that the most realistic value of E_F in the actual α -phase corresponds to the minimum of the pseudo-gap, i.e. $E_F - E_{F(\text{LMTO})} = -0.163$ eV for our calculation. Within a relaxation time approximation the diffusivity $D(E, \tau)$ is calculated. The D_B values (Fig. 2) are similar in magnitude to those obtained by Fujiwara *et al.* [14] for the idealized approximant $\alpha\text{-Al}_{114}\text{Mn}_{24}$ approximant. D_{NB} is almost independent on E , whereas the D_B values depend strongly on E and is particularly small in the pseudo-gap.

We present now the results of the conductivity calculations of the $\alpha\text{-AlMnSi}$ phase assuming the value of the Fermi energy given above i.e. $E_F - E_{F(\text{LMTO})} = -0.163$ eV. The Fig. 3 shows the predicted static conductivity (dc conductivity) versus the inverse scattering time. In the case of $\alpha\text{-Al}_{69.6}\text{Si}_{13.0}\text{Mn}_{17.4}$ two regimes

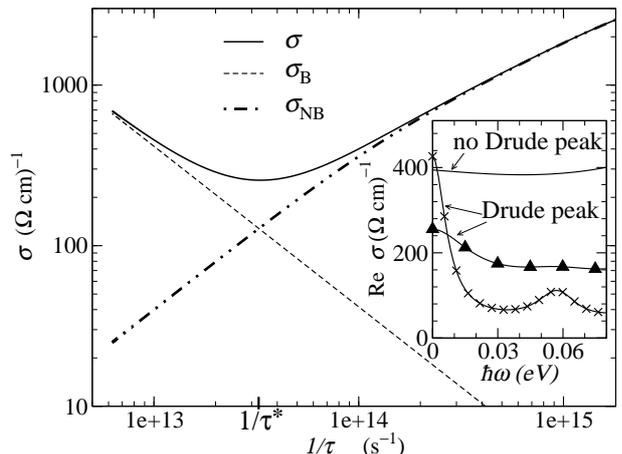


FIG. 3: Ab-initio dc conductivity, σ , in cubic approximant $\alpha\text{-Al}_{69.6}\text{Si}_{13.0}\text{Mn}_{17.4}$. σ is the sum of a Boltzmann term, σ_B , and a non Boltzmann term, σ_{NB} (see equation (8)). Inset: Real part, $\text{Re} \sigma(\omega)$, of the optical conductivity for three τ values. ω is the pulsation. Simple line, $\tau = \tau^*/3$; line with triangles, $\tau = \tau^* = 3.07 \cdot 10^{-14}$ s; line with crosses, $\tau = 3\tau^*$.

appear clearly: the metallic regime (Boltzmann regime) at large scattering time, $\tau > \tau^*$, and the insulating like regime (non Boltzmann regime) at small scattering time, $\tau < \tau^*$. $\tau^* = 3.07 \cdot 10^{-14}$ s is defined as the time for which the Boltzmann and non-Boltzmann contributions are equal. As expected from our model, σ_{NB} is almost proportional to $1/\tau$. Therefore, in the non Boltzmann regime, the conductivity increases with disorder as observed experimentally. For realistic τ values, $\tau < \tau^*$ [18], σ_{NB} dominates and σ increases when $1/\tau$ increases i.e. when defects or temperature increases. σ varies from $250 (\Omega \text{ cm})^{-1}$ for $\tau = 3.3 \cdot 10^{-14}$ s, to $2000 (\Omega \text{ cm})^{-1}$ for $\tau = 10^{-15}$ s. This is consistent with experimental results in $\alpha\text{-AlMnSi}$: $\sigma(4 \text{ K}) \simeq 200 (\Omega \text{ cm})^{-1}$ and $\sigma(900 \text{ K}) \simeq 2000 (\Omega \text{ cm})^{-1}$ and with standard estimates for the scattering time in these systems [5]. Furthermore for τ equals a few 10^{-14} s, i.e. when the Boltzmann term is negligible, the mean free path is given by the square root of the saturation value of ΔX_{NB}^2 and is of the order of 15 \AA . This is in agreement with estimates in the literature [5]. As discussed in [9] this means also that the systems is far from the Anderson transition despite its low conductivity. Within the relaxation time approximation used here, the optical conductivity is also the sum of two terms. One is the Boltzmann contribution (diagonal elements of the velocity operator) which gives rise to the so-called Drude peak and the other is the non Boltzmann conductivity (off diagonal elements of the velocity operator). As shown in the inset of Fig 3 a Drude peak can be identified in the Boltzmann regime, $\tau > \tau^*$, whereas in the non Boltzmann regime, $\tau < \tau^*$, the Drude peak disappears. This absence of the Drude peak is experimentally demonstrated for several icosahedral phases [19]. We note also that the physics of the non Boltzmann regime is similar to that obtained with anomalous diffu-

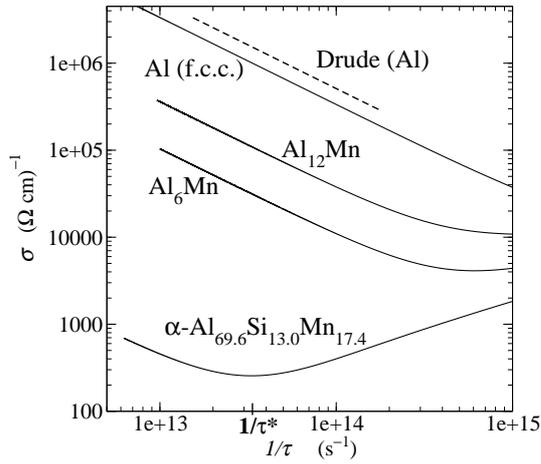


FIG. 4: Ab-initio electrical conductivity $\sigma(E_F)$ versus inverse scattering time $1/\tau$ (logarithmic scales): in Al, Al_{12}Mn , Al_6Mn , and in cubic approximant $\alpha\text{-Al}_{69.6}\text{Si}_{13.0}\text{Mn}_{17.4}$. In Al, the Boltzmann term dominates, and the model is compatible with a simple Drude model (dashed line) [1].

sion in the subdiffusive case [9, 20, 21, 22, 23].

Finally Fig. 4 exhibits the fundamental difference between the $\alpha\text{-AlMnSi}$ phase and a standard metal like Al (f.c.c.). Al_{12}Mn and Al_6Mn phases are somewhat intermediate and show the first signs of an insulating like regime at the strongest values of the scattering rate.

To conclude, a regime of charge carriers with small velocities has been identified, where quantum effects play a fundamental role and lead to a transition from a metallic like behavior to an insulating like behavior when the scattering rate increases. Quasicrystals and related complex metallic alloys give an example of this insulating like

regime. Other systems are known where the charge carriers velocity is small, either in all directions or in specific directions, thus we expect that the insulating like regime could be observed in other systems. For interacting electrons, a similar regime should also exist provided that the mean free path is smaller than a characteristic extension of the quasiparticle wavefunction, which means that the Fermi liquid picture breaks down. Several interacting electrons systems are good candidates for this small velocity regime. Recent results suggest this picture for a system like CeB_6 which is a heavy fermion system with large mass and thus small velocity. At low temperature T the Fermi liquid description is valid but at higher temperature ($T > T^*$) an incoherent insulating like regime is reached in which the resistivity decreases with increasing temperature. Within a Dynamical Mean Field Theory (DMFT) calculation [24] a disappearance of the Drude peak in the incoherent regime i.e. above T^* is predicted [6]. Recent theoretical results for the small polaron problem, with narrow polaronic band and thus small velocity, have been obtained also with the DMFT. They exhibit an analogous behavior [7]. The static resistivity (dc resistivity) increases with increasing temperature up to some T_0 and then decreases. Again in the incoherent regime above T_0 the Drude peak disappears [8]. A theoretical analysis similar to the discussion given here can be made also in the DMFT. It should give an additional insight in the quantum transport of correlated charge carriers with small velocities.

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