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THE ELECTRONIC RELAXATION TIME AND TRANSPORT PROPERTIES OF QUASICRYSTALS

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The novel model for electronic transport in quasicrystals is proposed. Assuming that the multi-valley fractional Fermi-surface exists in the icosahedral phases, the electronic relaxation time due to scattering by structural and phase disorder and quasilattice vibrations is calculated. Then we evaluate some kinetic characteristics like electrical conductivity and thermopower in a wide temperature range on the base of fractional Fermi surface model. The calculated transport properties reveal the main experimentally observed features of quasicrystals like low density of states on Fermi level, low electrical conductivity at zero temperature and its square-root temperature dependence at low temperatures, large absolute value and strong temperature dependence of thermopower.

The electronic transport in quasicrystals is one of the puzzling problems of the modern condensed matter physics and attracts a close attention of both theoreticians and experimentalists (for an extensive review see [1]). The fractal nature of Fermi surface and interplay between localization and delocalization of electronic states [2-5], the extremelly short electron mean free path, low carrier concentration and density of states on Fermi level, strong structural and phase disorder lead to the variety of speculations concerning the transport properties. However, since the electronic properties of quasicrystals exhibit still the metallic character (although with the features mentioned above), any theory of electronic transport should be based on some model of the metal-like electron spectrum with well-defined Fermi-surface. The main problem arising here is the effect of quasiperiodicity on the Fermi-liquid parameters. In the present paper we propose the simple model of electronic structure which accounts to a transport properties consistent with observed experimentally.

In the simplest Harrison approach to the construction of the Fermi surface the first approximation leads to the appearance of a number of electron voids on the places of the intersection of Harrison spheres and quasi-Brillouin zone boundaries and of hole voids in the icosahedrical angles. The following approximations lead to the subsequent splitting of these voids into smaller ones and so on. Because of icosahedral symmetry this splitting procedure continues indefinitely. As a result the volume of quasi-Brillouin zone as well as the size of each void tend to zero while the number of voids goes to infinity. In the limit the fractal structure appears. This situation takes place in an ideal quasicrystal, namely with no

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electron scattering processes and at zero temperature. Nevertheless in the real quasicrystal one has to take into account the smearing of the electron states in momentum space. Since the energy of quasiparticle is defined with in accuracy of its uncertainity $\delta\epsilon \sim \max\{T, \tau^{-1}\}$ (T is temperature, τ is the electron relaxation time), the splitting of Fermi surface within Harrison procedure has a sence as long as the characteristic size of voids is greater than $\delta\epsilon$. As a result we argue, that electronic structure of quasicrystal can be modeled as a set of electron and hole voids (or valleys) with the characteristic size of order of $\delta\epsilon$.

We consider N electron and hole voids distributed within the Brillouin zone. At zero temperature the finite relaxation time is solely due to structural disorder while with temperature increase the scattering on the quasilattice vibrations also gives rise to the electron relaxation. Let us introduce the effective chemical potential in each void (we use units in which $\hbar = k_B = c = 1$):

$$\mu_i = \frac{k_F^{(i)2}}{2m} \quad , \tag{1}$$

where $i = 1...N$ is the number and $k_F^{(i)}$ is the radius of a corresponding void and m is the effective mass of a charge carrier. First we consider the scattering of electron on structural imperfections. One can defines the Green function of electron as a $N \times N$ matrix:

$$G_{ij}(\mathbf{r}, \mathbf{r}', t, t') = - < T(\psi_i(\mathbf{r}, t)\psi_j^{\dagger}(\mathbf{r}', t') > \quad , \tag{2}$$

where ψ_i and $\psi_j^{\dagger}$ are electron field operators for i and j valleys. In the absence of scattering the $\hat{G}$ matrix is diagonal with

$$G_{ii}^{(0)}(\mathbf{k}, \omega_n) = \frac{1}{i\omega_n + \mu_i - (k^2/2m)}. \tag{3}$$

Scattering of electrons by structural imperfections results in both renormalization of diagonal elements and appearence of non-zero non-diagonal elements of $\hat{G}$ matrix [6]. We will consider the scattering to be isotropic and the corresponding amplitude $\hat{U}$ (which also is presented as $N \times N$ matrix) to be independent on the transfer momentum. Under these assumptions, the amplitudes of electron scattering between different valleys are the same, so $\hat{U} = U_0\hat{K}$, where U_0 is the amplitude for Born scattering of electron by impurity center and $K_{ij} = 1$ for all i and j. For the Green function averaged over the configuration of scattering centers we can write down the matrix Dyson equation:

$$\hat{G}^{-1} = [\hat{G}^{(0)}]^{-1} - \hat{\Sigma} \tag{4}$$

with $\hat{\Sigma}$ being the martix self-energy. The Dyson equation can be solved analytically in the case if all valleys have the same sizes: $\mu_i = \mu_0$ for all i. The details of calculation will be given elsewhere. In result the self-energy has the same matrix structure as $\hat{U}$: $\hat{\Sigma} = \Sigma_0\hat{K}$. The electron relaxation time is associated with Σ_0 as: $\tau^{-1} = 2\ i\ \mathrm{Im}\Sigma_0$. In result:

$$\frac{1}{\tau(\omega)} = \frac{N}{\tau_0}\left[1 + \frac{\omega}{\mu_0} + \left(\frac{N}{4\mu_0\tau_0}\right)^2\right]^{1/2} \tag{5}$$

In the last equation we have introduced the time $\tau_0^{-1} = n_{imp}|U_0|^2 m^{3/2}(2\mu_0)^{1/2}/\pi = n_{imp}|U_0|^2 \nu(\mu_0)/2\pi$ (n_{imp} is a concentration of scattering centers and $\nu(\mu_0)$ is the density of states on Fermi level). Finally one can conclude, that intervalley scattering results in the dramatic reducing of total relaxation time: $\tau \sim \tau_0/N$, where time τ_0 refers to intravalley scattering.

As temperature increases, the number of electrons scattered by quasilattice vibrations will increase with the corresponding reducing of total relaxation time. The electron-phonon interaction changes the electron momentum only by a quantity of the order of T/u, where u is the velocity of sound, while the momentum transfer of order of a^{-1} (a is the average interatomic distance in quasilattice) is needed to scatter electron from one valley to another. In this manner there exists a characteristic temperature: $T^* \sim u/a$ below which scattering by phonons is not able to provide effective intervalley scattering so only intravalley processes are possible. We argue that for temperatures $T < T^*$ the effective relaxarion time is given by:

$$\frac{1}{\tau_{tot}} = \frac{N}{\tau_0} + \frac{1}{\tau_{ph}(T)} \quad , \tag{6}$$

with $\tau_{ph}(T)$ being the electron relaxation time associated with electron-phonon scattering. In this temperature range the total electron relaxation is dominated by scattering on impurities even in pure samples due to large value of N. On the other hand if temperature exceeds over T^* the total relaxation time crosses over to regime with enhanced scattering by phonons. As a result, for $T > T^*$

$$\frac{1}{\tau_{tot}} = \frac{N}{\tau_0} + \frac{N}{\tau_{ph}(T)} \tag{7}$$

To understand the character of electrical conductivity one has to consider the parameter $k_F^{(0)}\ l \sim \mu_0\ \tau$ in comparison with unity (l is the electron mean free path). The value of this parameter governs whether the conductivity has metallic or non-metallic character. At zero temperature the size of the valley is $\mu_0 \sim \delta\epsilon \sim \tau_0^{-1}$. It means that condition of strong localization $\mu_0\ \tau_0 \sim 1$ is fulfilled independently on the disorder strength (even in the case of highly ordered icosahedral phase with $n_{imp} \to 0$). The physical reason for this phenomenon is the small momentum transfer $k_F^{(0)}$ involved in intravalley processes. Now we turn to intervalley processes. Here the momentum transfer Δk is of order of a^{-1}. In this case $\Delta k\ l \gg 1$. Hence, possibility of intervalley processes eliminates the localization situation and provides the non-zero (but small) conductivity of metallic character. The detailed theory which allows us to evaluate the intervalley conductivity within Kubo formalism shows [7] that for estimation of the residual conductivity one can use the Drude model with the effective relaxation time in the form of Eq. (5):

$$\sigma(T=0) = \sum_{i=1}^{N} \sigma_i \ \sim \ N\ e^2 v_F^{(0)2}\ \nu \frac{\tau_0}{N} \ \sim \ \frac{e^2 v_F^{(0)2}}{n_{imp}|U_0|^2}. \tag{8}$$

The magnitude of conductivity is extremely small due to its proportionality to $v_F^{(0)2}$. It is worth discussing the variation of $\sigma(T=0)$ with changing the strength of structural disorder. If the strength of disorder is close to zero, conductivity tends to zero, because neither intervalley nor intravalley processes are possible.

With the rise of n_{imp}, the non-zero $\sigma(T=0)$ appears, those magnitude could be calculated through Eq. (8). Thus we see, that residual conductivity should rise with the structural and phase disorder increase, as it is seen from experiments. We emphasize, that the reason for this effect is the possibility for electron to be scattered from one valley to another with the large momentum transfer of order of a^{-1}.

Now we turn to non-zero, but still low temperatures. In this case the intervalley scattering contribution is given by the same temperature independent Eq. (8). What concerns intravalley processes, they will contribute to conductivity in the same manner as temperature-dependent conductivity in the vicinity of metal-insulator transition. It is well established now [8], that at the edge of metal-insulator transition conductivity goes to zero according with $T^{1/2}$-law. Considering this contribution as a small correction to temperature-independent conductivity from intervalley processes, we can write at low temperatures:

$$\sigma(T) = \sigma(0) + \alpha \sqrt{T} \tag{9}$$

Under the further increase of temperature, the electron relaxation time will decrease due to scattering by phonons according to Eqs. (6) and (7). In this temperature range the contribution from intervalley processes begins to depend upon temperature, and total conductivity will be dominated by this contribution with the corresponding deviation from $T^{1/2}$-dependence.

Let us calculate the thermoelectric coefficient $\beta(T)$. We consider only intervalley scattering what is justified in the case of not too low temperature. One has:

$$\beta = \sum_{i=1}^{N} \beta_i \quad , \quad \beta_i = -\frac{1}{9}\pi^2 eT \frac{d}{d\mu_i}\left(v_i^{(0)2}\nu(\mu_i)\frac{\tau_0}{N}\right) \tag{10}$$

Simple estimate shows that:

$$\beta \sim -eTk_F^{(0)}\tau_0 \quad , \quad S = -\frac{\beta}{\sigma} \sim \frac{T}{e\mu_0} \quad , \tag{11}$$

where $S(T)$ is a thermopower (or Seebeck coefficient). As μ_0 is much less than the typical value of Fermi energies in metals, one concludes that diffusion thermopower of quasicrystals is much larger than in ordinary metals. Apart from the large absolute value of diffusion thermopower, the sign change with the temperature increase is also the characteristic feature of quasicrystals. Usually the thermopower sign change is attributed to the phonon processes which can account for the contributions opposite in sign to the diffusion thermopower [9]. These phonon contributions dominate over the diffusion component at temperatures $T \ll \Theta$ (Θ is Debye temperature), but are substentially reduced at $T \geq \Theta$ due to phonon-phonon scattering. To take into account the influence of electron-phonon interaction on thermopower one has to replace in Eq. (11) τ_0 by τ_{tot} given by Eqs. (6) and (7). The drag effect will affect thermopower at the temperature range between Θ^2/μ_0 and Θ. In this case [10]:

$$\beta \sim -e\left(\frac{T}{\Theta}\right)^2 \mu_0 k_F^{(0)}\tau_{tot} \quad , \quad S \sim -\frac{T^2}{e\Theta^2}. \tag{12}$$

Other feature of the thermopower in the model of fractional Fermi surface is the low value of degeneration temperature of electron gas. Indeed, electron gas could

be considered as Fermi gas if temperature is less than T_{deg} which is of order of magnitude μ_0 and in result can reach a hundred of Kelvins. It means that at $T > T_{deg}$ one has to consider electron gas as a Boltzman gas. In this case thermopower is nearly temperature independent and large in magnitude: $S \sim 1/e$ [11]. This effect also gives rise to unusually large absolute value of thermopower at high temperatures. The situation is complicated by the existence of two kinds of charge carriers. If the degeneration temperatures for electrons and holes differ, it is easy to understand that at some temperature between $T_{deg}^{(e)}$ and $T_{deg}^{(h)}$ thermopower should change a sign.

In comparison of our theory with experiment, one has to keep in mind that our model is of rather general character, so it is able to describe only a common features of quasicrystals distinctive from properties of usual metallic systems. Discussing experimental data the main problem is to reveal the properties unique to quasicrystals which arise solely from the quasiperiodicity.

Initial experimental studies of electron transport on the icosahedral phase indicated metallic-like behavior with large residual resistivities (of order 100 $\mu\Omega\ cm$) with the monotonic decrease of resistivity with temperature increasing up to room temperature [3]. Together with the strong temperature dependencies of thermopower and Hall coefficient, these observations were reminiscent of metallic-glass behavior [12]. However, further studies of Al-Cu-Fe(Ru) compounds able to form stable, nearly defect-free icosahedral phases, showed the enormally large resistivity up to 0.1 $\Omega\ cm$ [13] and even 1 $\Omega\ cm$ in system Al-Pd-Re [14], which increases with increasing of the degree of structural order. Clear, that these phases can not be considered by analogy with metallic glasses where structural disorder is the intrinsic property.

Different theoretical models were proposed to account for an unusual electron transport in quasicrystals, but all of them considered the case of ideal quasicrystal. As we pointed out before, in this case the Fermi surface has an infinite number of zero-area voids. Moreover, because of the special type of order, the electron wavefunctions can not be described by the Bloch form. Different theoretical approachs refered to ideal quasicrystals [8, 9] result in a controversial issues. Our model, applicable to real quasicrystals, provides the natural explanation for a low value of conductivity at zero temperature. Apart from conductivity, thermopower of quasicrystals exhibits enormous large absolute magnitude, which also can be considered as a unique property. Even ignoring possible phonon contributions, the large magnitude of electronic diffusion thermopower has also found explanation within our model due to two possible reasons: small size of valley and exteremely low degeneration temperature. It is easy to understand also the experimentally observed [3, 5] reducing of density of states on Fermi level with respect to the ordinary metals and dependence of electronic properties upon the degree of phase and structural ordering, because the effective Fermi energy and Fermi momentum are governed by the inverse electron relaxation time.

In summary, we have proposed the model of many-valley fractional Fermi-surface for electronic structure of quasicrystals which provides a natural explanation for a number of physical properties of these materials. The interplay between intravalley and intervalley scattering processes in combination with suggestion about the localization regime in each valley accounts for a small value of zero-temperature electrical conductivity and square-root temperature dependence at low temperatures, large absolute value and strong temperature dependence of thermopower.

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