Electronic relaxation time and transport properties of quasicrystals

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A novel model for electronic transport in quasicrystals is proposed. The electronic relaxation time due to scattering by structural and phase disorder and quasilattice vibrations is calculated on the assumption that a multivalley fractional Fermi surface exists in the icosahedral phases. Then some kinetic characteristics like the electrical conductivity and thermopower are evaluated over a wide temperature range on the basis of the fractional Fermi surface model. The calculated transport properties exhibit the main experimentally observed features of quasicrystals, such as the low density of states at the Fermi level, the low value of the electrical conductivity at zero temperature and its square-root temperature dependence at low temperatures, and the large absolute value and strong temperature dependence of the thermopower. © *1995 American Institute of Physics.*

The electronic transport in quasicrystals is one of the puzzling problems in modern condensed matter physics and attracts close attention from both theoreticians and experimentalists (for an extensive review see Ref. 1). The fractal nature of Fermi surface and the interplay between localization and delocalization of electronic states,[2–5] the extremely short electron mean free path, low carrier concentration and density of states at the Fermi level, strong structural and phase disorder lead to a variety of speculations concerning the transport properties. However, since the electronic properties of quasicrystals still exhibit a metallic character (although with the features mentioned above), any theory of electronic transport should be based on some model of a metal-like electron spectrum with a 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 a simple model of the electronic structure which leads to transport properties consistent with those 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 at points of inter-

section of the Harrison spheres and quasi-Brillouin zone boundaries and of hole voids on the icosahedral corners. The higher-order approximations lead to the subsequent splitting of these voids into smaller ones and so on. Because of the icosahedral symmetry this splitting procedure continues indefinitely. As a result, the volume of the 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 a fractal structure appears. This situation takes place in an ideal quasicrystal, namely, one with no electron scattering processes and at zero temperature. In a real quasicrystal, however, one has to take into account the smearing of the electron states in momentum space. Since the energy of a quasiparticle is defined within an uncertainty $\delta\epsilon \sim \max\{T, \tau^{-1}\}$ (T is the temperature, τ is the electron relaxation time), the splitting of the Fermi surface in the Harrison procedure has meaning as long as the characteristic size of the voids is greater than $\delta\epsilon$. As a result, we argue that electronic structure of a quasicrystal can be modeled as a set of electron and hole voids (or valleys) with a characteristic size of order $\delta\epsilon$.

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 as the temperature increases, the scattering on quasilattice vibrations also gives rise to electron relaxation. Let us introduce the effective chemical potential in each void (we use units in which $\hbar = k_B = c = 1$):

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

where $i = 1 \ldots N$ 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 an electron on structural imperfections. One can defines the Green function of electron as a $N \times N$ matrix:

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

where ψ_i and $\psi_j^{\dagger}$ are the electron field operators for valleys i and j. 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 a renormalization of the diagonal elements and the appearance of nonzero off-diagonal elements of the $\hat{G}$ matrix.[6] We will consider the scattering to be isotropic and the corresponding amplitude $\hat{\mathbf{U}}$ (which is also represented as an $N \times N$ matrix) to be independent of the momentum transfer. Under these assumptions, the amplitudes for electron scattering between different valleys are the same, and so $\hat{\mathbf{U}} = U_0 \hat{\mathbf{K}}$, where U_0 is the amplitude for Born scattering of an electron by an impurity center, and $K_{ij} = 1$ for all i and j. For the Green function averaged over configurations of the scattering centers we can write down the matrix Dyson equation:

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

where $\hat{\Sigma}$ is the self-energy matrix. The Dyson equation can be solved analytically in the case when all valleys have the same sizes: $\mu_i = \mu_0$ for all i. The details of the calculation will be given elsewhere. The result is that the self-energy has the same matrix structure as $\hat{\mathbf{U}}:\hat{\Sigma} = \Sigma_0 \hat{\mathbf{K}}$. The electron relaxation time is related to Σ_0 as: $\tau^{-1} = 2i \text{ Im } \Sigma_0$. Thus

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

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

As the temperature increases, the number of electrons scattered by quasilattice vibrations will increase, with a 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 a momentum transfer of the order of a^{-1} (a is the average interatomic distance in the 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 that only intravalley processes can occur. We argue that for temperatures $T < T^*$ the effective relaxation time is given by:

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

where $\tau_{\text{ph}}(T)$ is 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 the large value of N. On the other hand, if the temperature exceeds T^* the total relaxation time crosses over to a regime with enhanced scattering by phonons. As a result, for $T > T^*$

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

To understand the character of the 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 is of a metallic or nonmetallic character. At zero temperature the size of the valley is $\mu_0 \sim \delta\epsilon \sim \tau_0^{-1}$. This means that the condition of strong localization $\mu_0\tau_0 \sim 1$ is fulfilled independently of the degree of a disorder (even in the case of the highly ordered icosahedral phase with $n_{\text{imp}} \to 0$). 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 the order of a^{-1}. In this case $\Delta k l \gg 1$. Hence, the possibility of intervalley processes eliminates the localization situation and provides a nonzero (but small) conductivity of a metallic character. The detailed theory which allows us to evaluate the

intervalley conductivity within the Kubo formalism shows[7] that for estimation of the residual conductivity one can use the Drude model with an effective relaxation time in the form of Eq. (5):

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

The absolute value of the 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 degree of structural disorder. If the degree of disorder is close to zero, the conductivity tends to zero, because neither intervalley nor intravalley processes are possible. As $n_{\rm imp}$ is increased, a nonzero $\sigma(T=0)$ appears whose magnitude could be calculated through Eq. (8). Thus we see that residual conductivity should rise as the structural and phase disorder increase, as is seen in experiments. We emphasize that the reason for this effect is the possibility for an electron to be scattered from one valley to another with a large momentum transfer of the order of a^{-1}.

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

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

Upon a further increase in 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 the total conductivity will be dominated by this contribution, with a corresponding deviation from the $T^{1/2}$ dependence.

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

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

Simple estimates shows that

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

where $S(T)$ is the thermopower (or Seebeck coefficient). As μ_0 is much less than the typical values of the Fermi energy in metals, one concludes that the diffusion thermopower of quasicrystals is much larger than in ordinary metals. Besides the large absolute value of the diffusion thermopower, a change in sign as the temperature increases is also a characteristic feature of quasicrystals. Usually the thermopower sign

change is attributed to phonon processes which can give contributions opposite in sign to the diffusion thermopower.[9] These phonon contributions dominate over the diffusion component at temperatures $T \ll \Theta$ (Θ is the Debye temperature), but are substantially reduced at $T \geqslant \Theta$ due to phonon–phonon scattering. To take into account the influence of the electron–phonon interaction on the thermopower one has to replace τ_0 in Eq. (11) by τ_{tot} given by Eqs. (6) and (7). The drag effect will affect the thermopower in the temperature range between Θ^2/μ_0 and Θ. In this case[10]

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

Another feature of the thermopower in the fractional Fermi surface model is a low value of the degeneration temperature of the electron gas. Indeed, the electron gas can be regarded as a Fermi gas if the temperature is less than T_{deg}, which is of order of magnitude μ_0 and can therefore reach one hundred Kelvin. It means that at $T > T_{\text{deg}}$ one has to treat the electron gas as a Boltzmann gas. In this case the thermopower is nearly temperature independent and large in magnitude: $S \sim 1/e$ (Ref. 11). This effect also gives rise to an unusually large absolute value of the 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 $T_{\text{deg}}^{(e)}$ and $T_{\text{deg}}^{(h)}$ the thermopower should change sign.

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

Initial experimental studies of electron transport in the icosahedral phase indicated metal-like behavior with large residual resistivities (of the order of 100 $\mu\Omega\cdot$cm) with a monotonic decrease of the resistivity with increasing temperature up to room temperature.[3] Together with the strong temperature dependence of the 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 revealed an enormous resistivity, up to 0.1 $\Omega\cdot$cm (Ref. 13) and even 1 $\Omega\cdot$cm in the system Al–Pd–Re (Ref. 14) which increases with increasing degree of structural order. Clearly, these phases cannot be treated by analogy with metallic glasses, where the structural disorder is an intrinsic property.

Different theoretical models have been proposed to account for the unusual electron transport in quasicrystals, but all of them have considered the case of an 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 wave functions can not be described by the Bloch form. Different theoretical approaches pertaining to ideal quasicrystals[8,9] result in controversial issues. Our model, applicable to real quasicrystals, provides a natural explanation for the low value of the conductivity at zero temperature. Besides the conductivity, the enormous absolute magnitude of the thermopower of quasicrystals can also be regarded as a unique property. Even ignoring possible phonon contributions, the large magnitude of the electronic diffusion ther-

mopower has also found explanation within our model due to two possible causes: small valley size, and extremely low degeneration temperature. It is easy to understand also the experimentally observed[3,5] decrease in the density of states at the Fermi level as compared to ordinary metals and the dependence of the electronic properties on 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 a many-valley fractional Fermi-surface model for the 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 the suggestion of a localization regime in each valley accounts for the small value of the zero-temperature electrical conductivity and the square-root temperature dependence at low temperatures and the large absolute value and strong temperature dependence of the thermopower.

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