Anomalous \(c\)-axis transport in layered metals

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Electron transport in layered materials exhibits a number of unusual properties. The most striking example is a qualitatively different behavior of the in-plane (\(\rho_{ab}\)) and out-of-plane (\(\rho_c\)) resistivities: whereas the temperature dependence of \(\rho_{ab}\) is metallic-like, that of \(\rho_c\) is either insulating-like or even non-monotonic. At the level of non-interacting electrons, layered systems are metals with strongly anisotropic Fermi surfaces. A commonly used model is free motion along the planes and nearest-neighbor hopping between the planes:

\[
\varepsilon_k = \frac{k^2}{2m_{ab}} + 2J (1 - \cos k d),
\]

where \(k \parallel \) and \(k \perp \) are in the in-plane and \(c\)-axis components of momentum, respectively, \(m_{ab}\) is the in-plane mass, and \(d\) is lattice constant in the \(c\)-axis direction.

If the Hamiltonian consists of the band motion with spectrum \(\varepsilon_k\) and the interaction of electrons with potential disorder as well as with inelastic degrees of freedom, e.g., phonons, the Boltzmann equation predicts that the conductivities are given by

\[
\sigma_{ab} = e^2 \nu (v_{ab} \tau_{ab}), \quad \sigma_c = 4e^2 \nu J^2 d^2 \langle \sin^2 (k d) \rangle \tau_c,
\]

where \(\langle \ldots \rangle\) denotes averaging over the Fermi surface and over the thermal (Fermi) distribution, \(\nu = m_{ab}/\pi d\) is the density of states, and \(\tau_{ab}\) is the transport time, resulting from all scattering processes (we set \(\hbar = k_B = 1\)). If \(\tau_c\) decreases with the temperature, both \(\sigma_{ab}\) and \(\sigma_c\) are expected to decrease with \(T\) as well. This is not what the experiment shows.

The \(c\)-axis puzzle received a lot of attention in connection to the HTC materials, and a non-Fermi-liquid nature of these materials was suggested to be responsible for the anomalous \(c\)-axis transport. However, other materials, such as graphite, TaS\(_2\), Sr\(_2\)RuO\(_4\), organic metals, etc., behave as canonical Fermi liquids in all aspects but the \(c\)-axis transport. This suggests that the origin of the effect is not related to the specific properties of HTC compounds but common for all layered materials. A large number of models were proposed to explain the \(c\)-axis puzzle. Despite this variety, most authors seem to agree on that the coherent band transport in the \(c\)-axis direction is destroyed. Although there is no agreement as to what replaces the band transport in the "incoherent" regime, the most frequently discussed mechanisms include incoherent tunneling between the layers, assisted by either out-of-plane impurities or by coupling to dissipative environment or polarons.

The message of this Letter is two-fold. First, we observe that neither elastic nor inelastic (electron-phonon) scattering can destroy band transport even in a strongly anisotropic metal as long as the familiar parameter \(E_F \tau\) is large. Nothing happens to the Boltzmann conductivities in Eq.\(2\) except for \(\sigma_c\) becoming very small at high temperatures so that other mechanisms, not included in Eq.\(2\), dominate transport. This observation is in agreement with recent experiment where a coherent feature (angle-dependent magnetoresistance) was observed in a supposedly incoherent regime. Second, we propose phonon-assisted tunneling through resonant impurities as the mechanism competing with the band transport. As such tunneling provides an additional channel for trans-
port, the total conductivity is
\[ \sigma_c = \sigma_c^B + \sigma_{\text{res}}, \]
where \( \sigma_{\text{res}} \) is the resonant-impurity contribution. Because \( \sigma_{\text{res}} \) increases with the temperature, the band channel is short-circuited by the resonant one at high enough temperatures\[5\]. Accordingly, \( \sigma_c \) goes through a minimum at a certain temperature (and \( \rho_c = \sigma_c^{-1} \) goes through a maximum). We consider phonon-assisted tunneling through a wide band of resonant levels distributed uniformly in space. We show that the non-perturbative (in the electron-phonon coupling) version of this theory is in a quantitative agreement with the experiment on Sr$_2$RuO$_4$\[5\]. Due to a similarity between phonon-assisted tunneling and other problems, in which interaction leads to the formation of a cloud surrounding the nucleation through a wide band of resonant levels distributed simultaneously in all directions\[16, 17, 18\] and maximum at a certain temperature (and \( \rho_c = \sigma_c^{-1} \) goes through a maximum). We consider phonon-assisted tunneling and provides a microscopic theory for some of the mechanisms considered in prior work. We begin with the discussion of the breakdown (or lack of it thereof) of the Boltzmann equation.

One may wonder whether the band transport along the c-axis breaks down because the Anderson localization occurs in the c-direction whereas the in-plane transport remains metallic. This does not happen, however, because an electron, encountering an obstacle for motion along the c-axis, moves quickly to another point in the plane, where such an obstacle is absent. More formally, it has been shown the Anderson transition occurs only simultaneously in all directions\[12, 13, 14, 15\] and only if \( J \) is exponentially smaller than 1/\( \tau \). Therefore, localization cannot explain the observed behavior.

Refs.\[12, 20\] suggested an idea of the “coherent-incoherent crossover”. It implies that the coherent band motion breaks down if electrons are scattered faster than they tunnel between adjacent layers, i.e., if \( J \tau \ll 1 \). Consequently, the current in the c-direction is carried via incoherent hops between conducting layers. It was noted by a number of authors that the assumption about incoherent nature of the transport does not, by itself, explain the difference in temperature dependences of \( \sigma_{ab} \) and \( \sigma_c \).\[20, 21\]. Due to conservation of the in-plane momentum, \( \sigma_c \) is proportional to \( \tau \) both in the coherent and incoherent regimes. Nevertheless, an issue of the “coherent-incoherent crossover” poses a fundamentally important question: can scattering destroy band transport only in some directions, if the spectrum is anisotropic enough? We argue here that this is not the case.

Since we have already ruled out elastic scattering, this leaves inelastic one as a potential culprit. We focus on the case of the electron-phonon interaction as a source of inelastic scattering. For an isotropic metal, the quantum kinetic equation is derived from the Keldysh equations of motion for the Green’s function via the Prange-Kadanoff procedure\[22\] for any strength of the electron-phonon interaction. In this Letter, we apply the Prange-Kadanoff theory to metals with strongly anisotropic Fermi surfaces, such as the one in Fig. 1. We show that, exactly as in the isotropic case, the Boltzmann equation holds its standard form as long as \( E_F \tau_{\text{ep}} \gg 1 \). Since this form does not change between coherent (\( J \tau_{\text{ep}} \gg 1 \)) and incoherent (\( J \tau_{\text{ep}} \ll 1 \)) regimes, it means that the coherent-incoherent crossover is, in fact, absent.

We adopt the standard Fröhlich Hamiltonian for the deformation-potential interaction with longitudinal acoustic phonons \( (\omega_q = \omega) \)
\[
H = \sum_k \epsilon k a_k^+ a_k + \sum_q \omega_q b_q^+ b_q + \sum_{k,q} g_q \sqrt{\omega_q} d_{k+q}^+ a_k b_{q-k} + b_{q-k}^+ a_k, \tag{4}
\]
Since tunneling matrix elements are much more sensitive to the increase in the inter-plane distance than the elastic moduli, the anisotropy of phonon spectra in layered materials, albeit significant, is still weaker than the anisotropy of electron spectra (see, e.g., Ref. \[24\]). Therefore, we treat phonons in the isotropic approximation, and assume that the magnitude of the Fermi velocity is larger than the speed of sound \( s \).

For a static and uniform electric field, the Keldysh component of the electron’s Green function satisfies the Dyson equation
\[
\hat{L} G^K + \frac{i}{2} \left( [\text{Re} \Sigma^K, \otimes G^K]_+ + [\Sigma^K, \otimes \text{Re} G^K]_- \right) = \frac{1}{2} \left( [\Sigma^K, \otimes A]_+ + [\Gamma, \otimes G^K]_+ \right). \tag{5}
\]
Here \( \hat{L} = (\partial_t + v \cdot \nabla R + eE \cdot \nabla k) \) is the Liouville operator, \( A = i (G^K - G^A) \) is the spectral function, \( \Gamma = i (\Sigma^K - \Sigma^A) \), and \( \otimes \) denotes the convolution in space and time. Thanks to the Migdal theorem, the self-energy does not change between coherent (\( J \tau_{\text{ep}} \gg 1 \)) and incoherent (\( J \tau_{\text{ep}} \ll 1 \)) response, when the self-energy
\[
\hat{L} g^K + \frac{i}{2} [\text{Re} \Sigma^K, g^K]_- = 2i \Sigma^K - \frac{1}{2} [\Gamma, g^K]_+ \tag{6}
\]
for the “distribution function”
\[
g^K(\epsilon, \hat{n}) = \frac{i}{\pi} \int G^K(\epsilon, \xi_k, \hat{n}) d\xi_k, \tag{7}
\]
where \( \hat{n} = v_k / |v_k| \) is a local normal to the Fermi surface.

We consider a linear dc response, when the self-energy is needed only at equilibrium. Within the Migdal theory, the Matsubara self-energy is given by a single diagram
\[
\Sigma(\epsilon, \hat{n}) = -\int \frac{d\omega}{2\pi} \int \frac{d^3q}{(2\pi)^3} g^2(q) G(\epsilon - \omega, k - q) D(\omega, q),
\]
where the dressed phonon propagator
\[ D^{-1} = D_0^{-1} - g^2 \Pi \]
is expressed through bare one
\[ D_0(\omega, q) = -s^2 q^2 / (\omega^2 + s^2 q^2) \]
and polarization operator \( \Pi \) which, for \( E_F > 2J \), is given by its 2D form
\[ \Pi(\omega, q) = -\nu \left( 1 - |\omega|/\sqrt{v_F^2 q^2 + \omega^2} \right). \]

We assume that the electron-phonon vertex decays on some scale \( k_D \) shorter than Fermi momentum \( (k_D \ll k_F) \). This assumption allows one to linearize the dispersion \( \xi_k - q \approx \xi_k - v_F \cdot q \) and simplifies the analysis without changing the results qualitatively. As long as \( J \ll E_F \), we have \( |v_k| \approx k_F/m_{ab} \approx v_F \), where \( k_F \) is the radius of the cylinder in Fig. 1 for \( J = 0 \). Despite the fact that the electron velocity does have a small component along the c-axis, its in-plane component is large (cf. Fig. 1). Since it is the magnitude of \( v_k \) that controls the Migdal’s approximation, the problem reduces to the interaction of fast 2D electrons with slow 3D phonons. With these simplifications, we find
\[
\begin{align*}
\text{Re} \Sigma^R(\epsilon, \tilde{n}) &= -\frac{1}{4} \frac{\zeta}{1 - \zeta} \left( \frac{k_D}{k_F} \right)^2 \epsilon; \quad \text{(8a)} \\
\text{Im} \Sigma^R(\epsilon, \tilde{n}) &= -\frac{\zeta^2}{12(1 - \zeta)^2} \omega_D^2, \quad \text{(8b)}
\end{align*}
\]
where \( \zeta = \nu \gamma^2 \) is a dimensionless coupling constant and \( \omega_D = sk_D \). We see that, despite the strong anisotropy, the self-energy remains local, i.e., independent of \( \xi_k \).

Vertex renormalization leads to two types of corrections to the self-energy: those that are proportional to the Migdal’s parameter \( (s/v_F) \) and those that are proportional to \( ms^2/\epsilon \). The second type of corrections invalidates the Migdal’s theory for temperatures below \( ms^2 \), which is about 1 K in a typical metal. For metals with anisotropic spectrum the existence of such a scale is potentially dangerous, since it is not obvious which of the masses (light or heavy) defines this scale. We find that the in-plane mass \( (m_{ab}) \) controls the vertex renormalization for the nearly cylindrical Fermi surface. This shows that the Migdal theory for layered metals has the same range of applicability as for isotropic metals [25].

The rest of the derivation proceeds in the same way as for the isotropic case [26], and the resulting Boltzmann equation assumes its standard form. Since no assumption about the relation between \( \tau_{e-\text{ph}} \) and the dwell time \( (1/J) \) has been made, the conductivities obtained from the Boltzmann equation have the same form regardless of whether \( J \tau_{e-\text{ph}} \) is large or small. In other words, there is no coherent-incoherent crossover due to inelastic scattering in an anisotropic metal [29].

The situation changes qualitatively if resonant impurities are present in between the layers. Electrons that tunnel through such impurities are moving with the speed controlled by the broadening of a resonant level, i.e., much slower than speed of sound. For that reason they can not be treated within the formalism outlined above and require a separate study.

To evaluate the resonant-impurity contribution to the conductivity, we assume that the impurities are randomly distributed in space with density \( n_{imp} \), whereas their energy levels uniformly distributed over an interval \( E_b \). The tunneling conductance of a bilayer junction is
\[
G = e^2 \int d\epsilon d\epsilon' W_{\epsilon, \epsilon'} \left[ \frac{\partial n_e(1 - n'_e)}{\partial \epsilon} + \frac{\partial n'_e}{\partial \epsilon'} n_e \right], \quad \text{(9)}
\]
where \( W_{\epsilon, \epsilon'} \) is a transition probability per unit time and \( n_e \) is the Fermi function. To calculate \( W_{\epsilon, \epsilon'} \), we use the results of Ref. [31] for the probability of phonon-assisted tunneling through a single impurity
\[
W_{\epsilon, \epsilon'} = \Gamma_L \Gamma_R \int_{-\infty}^{\infty} dt_1 e^{it_1} \left\{ \int_0^{\infty} dt_2 dt_3 e^{i(t_2 - t_3)(\epsilon - \bar{\epsilon}_0) - \Gamma(t_2 + t_3)} \right\} \times \exp \left( -\sum_q \frac{|\alpha_q|^2}{2\omega_q^2} \left[ 1 - e^{-i\omega_q} + e^{i\omega_q} (e^{-i\omega_q} - 1) \right]^2 \coth \left( \frac{\omega_q}{2T} \right) + (e^{-i\omega_q} + e^{i\omega_q} + e^{i\omega_q} (e^{-i\omega_q} - 1)(1 - e^{i\omega_q}) - c.c. \right) \right),
\]
produces the well-known Breit-Wigner formula. From now on, we consider a wide band of resonant levels: \( E_b \gg T \gg \Gamma \). Averaging Eq. (10) over spatial and en-
ergy positions of resonant levels, one obtains

\[
\sigma_{\text{res}} = \sigma_0 \int_{-\infty}^{\infty} \frac{d\epsilon}{2\pi} \left[ \coth \left( \frac{\epsilon}{2T} \right) + \frac{1}{2T \sinh^2 \left( \frac{\epsilon}{2T} \right)} \right] \int_{-\infty}^{\infty} dt e^{-\lambda f(t)}
\]

\[
f(t) = \int_{0}^{\omega_D} d\omega \frac{\omega}{\omega_D} \left( (1 - \cos(\omega t)) \coth \left( \frac{\omega}{2T} \right) + i \sin(\omega t) \right).
\]

Here \( \sigma_0 \) is the conductivity due to elastic resonant tunneling and \( \lambda \equiv \Lambda^2 \omega_D^2 / \rho s^2 \pi^2 \) is the dimensionless coupling constant for localized electrons. In the absence of electron-phonon interaction, \( \sigma_{\text{res}} \) is temperature independent and given by \( \sigma_{\text{res}} \simeq \pi e^2 \Gamma_1 n_{\text{imp}} a_0 d / E_D \), where \( a_0 \) is the localization radius of a resonant state and \( \Gamma_1 \simeq \omega_0 e^{-d/a_0} \) is its typical width. We note that the electron-phonon interaction is much stronger for localized electrons than for band ones: \( \lambda / \zeta \sim (k_F d) (v_F / s) \gg 1 \).

Since typically \( \zeta \sim 1 \), one needs to consider a non-perturbative regime of phonon-assisted tunneling. In that case, resonant tunneling is exponentially suppressed at \( T = 0 \): \( \sigma_{\text{res}}(T = 0) = \sigma_0 e^{-\lambda/2} \). At finite \( T \), we find

\[
\sigma_{\text{res}} = \sigma_0 e^{-\lambda/2} \left( 1 + \frac{\pi \lambda}{3} \left( \frac{T}{\omega_D} \right)^2 \right), \quad T \ll \frac{\omega_D}{\sqrt{\lambda}}.
\]

As \( T \) increases, \( \sigma_{\text{res}} \) grows, resembling the zero-bias anomaly in disordered metals and Mössbauer effect. At high temperatures \( (T \gg \omega_D) \), \( \sigma_{\text{res}} \) approaches the non-interacting value \( \sigma_0 \). The asymptotic regimes in the interval \( \omega_D / \sqrt{\lambda} \ll T \ll \omega_D \) can also be studied but we will not pause for this here. Notice, that in contrast to the phenomenological model of Ref. [8], there is no simple relation between the \( T \)-dependencies of \( \sigma_c^B \) and \( \sigma_{\text{res}} \).

To compare our model with the experiment, we extract \( \sigma_c^B \) from the low-temperature (between 10 and 50 K) \( c \)-axis resistivity of \( \text{Sr}_2\text{RuO}_4 \) and extrapolate it to higher temperatures [2]. The resonant part of the conductivity is calculated numerically using Eq. (11). The fit to the data for \( \sigma_{\text{cl}} = 4.3 \times 10^{12} \Omega^{-1} \text{cm}^{-1}, \omega_D = 41 \text{K} \) and \( \lambda = 16 \) is shown in Fig. 2. The agreement between the theory and experiment is quite good and the values of the fitting parameters are reasonable. An immediate consequence of our model is the sample-to-sample variation of the \( c \)-axis conductivity. Among the layered materials, the largest amount of data is collected for graphite [2]. Even within the group of samples with comparable in-plane mobilities, the temperature of the maximum in \( \rho_c \) varies from 40K to 300 K [2, 23].

To conclude, we have shown that the Boltzmann equation and its consequences are no less robust for anisotropic metals than they are for isotropic ones. The only condition controlling the validity of the Boltzmann equation is the large value of \( E_F \tau \), regardless of whether \( \tau \) comes from elastic or inelastic scattering. Out-of-plane localized states change the \( c \)-axis transport radically while playing only minor role for the in-plane one. While \( \rho_{ab} \) remains metallic, an interplay between phonon-assisted tunneling and conventional momentum relaxation causes insulating or non-monotonic dependence of \( \rho_c \) on temperature. This model is in a good agreement with the experimental data on \( \text{Sr}_2\text{RuO}_4 \).

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