Anomalous c-Axis Transport in Layered Metals

D. B. Gutman and D. L. Maslov

Department of Physics, University of Florida, Gainesville, Florida 32611, USA

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Transport in metals with a strongly anisotropic single-particle spectrum is studied. Coherent band transport in all directions, described by the standard Boltzmann equation, is shown to withstand both elastic and inelastic scattering as long as $E_F\tau \gg 1$. A model of phonon-assisted tunneling via resonant states located in between the layers is suggested to explain a nonmonotonic temperature dependence of the c-axis resistivity observed in experiments.

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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 (ρ_{ab}) and out-of-plane (ρ_c) resistivities: whereas the temperature dependence of ρ_{ab} is metallic-like, that of ρ_c is either insulating-like or even nonmonotonic. At the level of noninteracting 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:

$$\varepsilon_{\mathbf{k}} = \mathbf{k}_{||}^2/2m_{ab} + 2J(1 - \cos k_\perp d), \tag{1}$$

where $\mathbf{k}_{||}$ 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. For the strongly anisotropic case ($J \ll E_F$), the equipotential surfaces are ''corrugated cylinders.''

If the Hamiltonian consists of the band motion with spectrum (1) 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

$$\sigma_{ab}^B = e^2\nu\langle v_a v_b \tau_{\mathrm{tr}}\rangle, \qquad \sigma_c^B = 4e^2\nu J^2 d^2\langle \sin^2(k_\perp d)\tau_{\mathrm{tr}}\rangle, \tag{2}$$

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 τ_{tr} is the transport time, resulting from all scattering processes (we set $\hbar = k_B = 1$). If τ_{tr} decreases with the temperature, *both* σ_{ab} and σ_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 high T_c materials [1], and a non-Fermi-liquid nature of these materials was suggested to be responsible for the anomalous c-axis transport [2]. However, other materials, such as graphite [3], TaS_2 [4], Sr_2RuO_4 [5], organic metals [6], 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 high T_c 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 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 interplane impurities [7–10] or by coupling to dissipative environment [11], and polarons [12,13]. Recent experiment [14] has shown that the c-axis resistivity is reduced by introducing defects into a clean organic metal κ-(BEDT-TTF)$_2$-Cu(SCN)$_2$. This observation gives strong support to the scenario of transport through interplane impurities.

The message of this Letter is twofold. First, we observe that neither elastic or 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 σ_c^B becoming very small at high temperatures so that other mechanisms, not included in Eq. (2), dominate transport. Second, we propose tunneling through resonant impurities as the mechanism competing with the band transport. As such tunneling provides an additional channel for transport, the total conductivity is [7,14,15]

$$\sigma_c = \sigma_c^B + \sigma_{\mathrm{res}}, \tag{3}$$

where σ_{res} is the resonant-impurity contribution. If σ_{res} increases with T, the band channel is short-circuited by the resonant one at high enough temperatures. Accordingly, $\rho_c = \sigma_c^{-1}$ goes through a minimum at a certain temperature. There are a number of mechanisms that can lead to an increase of σ_{res} with T, e.g., the energy dependence of the transmission coefficient. Here, we focus on a generic mechanism of phonon-assisted tunneling through a wide band of resonant levels and illustrate the feasibility of this mechanism for two materials: Sr_2RuO_4 [5] and κ-(BEDT-TTF)$_2$-Cu(SCN)$_2$ [14].

Because of a similarity between phonon-assisted tunneling and other problems, in which interaction leads to the formation of a cloud surrounding the electron (such as

polaronic effect and zero-bias anomaly), many ideas put forward earlier [7–13] agree with our picture. Nevertheless, we believe that only a combination of resonant impurities and electron-phonon interaction solves the puzzle of c-axis resistivity 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 transition occurs in the c-direction whereas the in-plane transport remains metallic. This does not happen, however, because Anderson localization occurs either simultaneously in all directions [for $J < \tau^{-1}\exp(-E_F\tau)$] or in none [16–18]. On a more intuitive level, quantum interference—the cause of localization—for trajectories along the c-axis is destroyed by the in-plane motion, while interference of in-plane trajectories is destroyed by the motion in the c-direction. Therefore, as long as an electron remains delocalized in any of these directions, it remains delocalized in others as well, and Anderson localization cannot explain the observed behavior.

References [19,20] 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 σ_{ab} and σ_c [20,21]: due to conservation of the in-plane momentum, σ_c is proportional to τ 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 [22,23]? We argue here that this is not the case.

Since we have already ruled out elastic scattering, this leaves the 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 [24] for any strength of the electron-phonon interaction.

In this Letter, we apply the Prange-Kadanoff theory to metals with anisotropic Fermi surfaces. We show that, exactly as in the isotropic case, the Boltzmann equation holds its standard form as long as $E_F\tau_{\text{e-ph}} \gg 1$. Since this form does not change between coherent ($J\tau_{\text{e-ph}} \gg 1$) and incoherent ($J\tau_{\text{e-ph}} \ll 1$) regimes, it means that the coherent-incoherent crossover is, in fact, *absent*.

We adopt the standard Frölich Hamiltonian for the deformation-potential interaction with longitudinal acoustic phonons ($\omega_q = sq$)

$$H = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} a^\dagger_{\mathbf{k}} a_{\mathbf{k}} + \sum_{\mathbf{q}} \omega_q b^\dagger_{\mathbf{q}} b_{\mathbf{q}} + \sum_{\mathbf{k},\mathbf{q}} g_q \sqrt{\omega_q}\, a^\dagger_{\mathbf{k}+\mathbf{q}} a_{\mathbf{k}} (b_{\mathbf{q}} + b^\dagger_{-\mathbf{q}}). \quad (4)$$

Since tunneling matrix elements are much more sensitive to an increase in the interplane distance than 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. [25]). 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

$$2\hat{L}G^K + i[\mathrm{Re}\Sigma^R, G^K] + i[\Sigma^K, \mathrm{Re}G^K] = \{\Sigma^K, A\} - \{\Gamma, G^K\}. \quad (5)$$

Here, $\hat{L} = (\partial_t + \mathbf{v}\cdot\nabla_{\mathbf{R}} + e\mathbf{E}\cdot\nabla_{\mathbf{k}})$ is the Liouville operator, and $A = i(G^R - G^A)$ is the spectral function, $\Gamma = i(\Sigma^R - \Sigma^A)$. Thanks to the Migdal theorem, the self-energy does not depend on electron's dispersion $\xi_{\mathbf{k}} \equiv \varepsilon_{\mathbf{k}} - E_F$, and Eq. (5) can be integrated over $\xi_{\mathbf{k}}$. This results in an equation

$$\hat{L}g^K + \frac{i}{2}[\mathrm{Re}\Sigma^R, g^K]_- = 2i\Sigma^K - \frac{1}{2}\{\Gamma, g^K\} \quad (6)$$

for the "distribution function" $g^K(\epsilon, \hat{n}) = (i/\pi)\times \int G^K(\epsilon, \xi_{\mathbf{k}}, \hat{n}) d\xi_{\mathbf{k}}$, where $\hat{n} = \mathbf{v}_{\mathbf{k}}/|\mathbf{v}_{\mathbf{k}}|$.

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

$$\Sigma(\epsilon, \hat{n}) = -\int \frac{d\omega}{2\pi} \int \frac{d^3q}{(2\pi)^3} g^2(q) G(\epsilon - \omega, \mathbf{k} - \mathbf{q}) D(\omega, q),$$

where the dressed phonon propagator, $D^{-1} = D_0^{-1} - g^2\Pi$, is expressed through the bare one, $D_0(\omega, q) = -s^2q^2/(\omega^2 + s^2q^2)$, and polarization operator Π which, for $E_F > 2J$, is given by its 2D form $\Pi(\omega, q) = -\nu(1 - |\omega|/\sqrt{v_F^2 q_\parallel^2 + \omega^2})$. 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 $\xi_{\mathbf{k}-\mathbf{q}} \approx \xi_{\mathbf{k}} - \mathbf{v}_{\mathbf{k}}\cdot\mathbf{q}$ and simplifies the analysis without changing the results qualitatively. As long as $J \ll E_F$, we have $|\mathbf{v}_{\mathbf{k}}| \approx k_F/m_{ab} \approx v_F$, where k_F is the radius of the Fermi surface 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. Since it is the magnitude of $\mathbf{v}_{\mathbf{k}}$ 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

$$\Sigma^R(\epsilon, \hat{n}) = -\frac{1}{4}\frac{\zeta}{1-\zeta}\left(\frac{k_D}{k_F}\right)^2 \epsilon - i\frac{\zeta}{12(1-\zeta)^2}\frac{|\epsilon|^3}{\omega_D^2} \quad (7)$$

where $\zeta = \nu g^2$ 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 $\xi_{\mathbf{k}}$.

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/ϵ. 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 [26].

The rest of the derivation proceeds in the same way as for the isotropic case [24], and the resulting Boltzmann equation assumes its standard form. Since no assumption about the relation between $\tau_{\text{e-ph}}$ and the dwell time $(1/J)$ has been made, the conductivities obtained from the Boltzmann equation have the same form regardless of whether $J\tau_{\text{e-ph}}$ is large or small. In other words, there is no coherent-incoherent crossover due to inelastic scattering in an anisotropic metal [28].

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

$$G = -e^2 \int d\epsilon d\epsilon' W_{\epsilon,\epsilon'} \left[\frac{\partial n_\epsilon}{\partial \epsilon}(1 - n_{\epsilon'}) + \frac{\partial n_{\epsilon'}}{\partial \epsilon'} n_\epsilon \right], \quad (8)$$

where $W_{\epsilon,\epsilon'}$ is a transition probability per unit time and n_ϵ is the Fermi function. For a single impurity [29,30],

$$W_{\epsilon,\epsilon'} = \Gamma_L \Gamma_R \int_{-\infty}^{\infty} dt_1 e^{it_1(\epsilon'-\epsilon)} \int_0^{\infty} dt_2 dt_3 e^{i(t_2-t_3)(\epsilon-\bar{\epsilon}_0)-\Gamma(t_2+t_3)} \times \exp\left(-\sum_q \frac{|\alpha_q|^2}{2\omega_q^2} \left[|1 - e^{-i\tau_3} + e^{i\tau_1}(e^{-i\tau_2} - 1)|^2 \coth\left(\frac{\omega_q}{2T}\right) + [e^{-i\tau_3} + e^{i\tau_2} + e^{i\tau_1}(e^{-i\tau_2} - 1)(1 - e^{i\tau_3}) - \text{c.c.}] \right] \right), \quad (9)$$

where $\tau_i = \omega_q t_i$ $(i = 1 \ldots 3)$, $\alpha_q = -i\Lambda q/\sqrt{\rho\omega_q}$, Λ is the deformation-potential constant, Γ_L and Γ_R are tunneling widths of the resonant level, $\Gamma = \Gamma_L + \Gamma_R$, and $\bar{\epsilon}_0$ is the energy of a resonant level renormalized by the electron-phonon interaction. In the limit of no electron-phonon interaction, Eq. (9) reproduces the Breit-Wigner formula. From now on, we consider a wide band of resonant levels: $E_b \gg T \gg \Gamma$. Averaging Eq. (9) over spatial and energy positions of resonant levels, we obtain

$$\sigma_{\text{res}} = \sigma_{\text{el}} \int_{-\infty}^{\infty} dt \frac{i\pi T^2 t}{\sinh^2(\pi T t + i0)} e^{-\lambda f(t)}$$
$$f(t) = \int_0^{\omega_D} d\omega \frac{\omega}{\omega_D^2} \left[[1 - \cos(\omega t)] \coth\left(\frac{\omega}{2T}\right) - i \sin(\omega t) \right]. \quad (10)$$

Here, σ_{el} is the conductivity due to elastic resonant tunneling, and $\lambda \equiv \Lambda^2 \omega_D^2/\rho s^5 \pi^2$ is the dimensionless coupling constant for localized electrons. In the absence of electron-phonon interaction, σ_{res} is temperature independent and given by $\sigma_{\text{el}} \simeq \pi e^2 \Gamma_1 n_{\text{imp}} a_0 d/E_b$ [31], where a_0 is the localization radius of a resonant state and $\Gamma_1 \propto \epsilon_0 p_F e^{-d/a_0}/(p_F^2 + a_0^{-2})d$ is its typical width. We note that the electron-phonon interaction is much stronger for localized electrons than for band ones: $\lambda/\zeta \sim (k_F d) \times (v_F/s) \gg 1$. Since typically $\zeta \sim 1$, one needs to consider a nonperturbative regime of phonon-assisted tunneling. In that case, resonant tunneling is exponentially suppressed at $T = 0$: $\sigma_{\text{res}}(T = 0) = \sigma_{\text{el}} e^{-\lambda/2}$. At finite T, we find

$$\sigma_{\text{res}} = \sigma_{\text{el}} \begin{cases} e^{-\lambda/2}[1 + \frac{\pi^2\lambda}{3}(\frac{T}{\omega_D})^2], & T \ll \frac{\omega_D}{\sqrt{\lambda}}, \\ 1 - \frac{\lambda}{9}\frac{\omega_D}{T}, & T \gg \lambda\omega_D. \end{cases}$$

As T increases, σ_{res} growth, resembling the zero-bias anomaly in disordered metals and Mössbauer effect. At high temperatures ($T \gg \lambda\omega_D$), σ_{res} approaches the noninteracting value (σ_{el}). The asymptotic regimes in the interval $\omega_D/\sqrt{\lambda} \ll T \ll \lambda\omega_D$ can also be studied, but we will not pause for this here. Depending on the parameters of the Boltzmann and tunneling parts of the conductivity, the total resistivity may exhibit a variety of T dependences: purely metallic (for weak tunneling), purely insulating (for strong tunneling), minimum at low T, maximum at high T, and both minimum and maximum. All of this behaviors are observed in the experiment. For example, ρ_c (i) is purely metallic in overdoped high T_c cuprates; (ii) has a minimum in underdoped ones [1]; (iii) is purely insulating in TaS_2 [4]; (iv) has both a minimum and a maximum in graphite [32]; and (v) has a maximum in Sr_2RuO_4 [5], κ-(BEDT-TTF)$_2$-Cu(SCN)$_2$ [14], $(Bi_{0.5}Pb_{0.5})_2Ba_3Co_2O_y$ and $NaCo_2O_4$ [23]. Deferring a detailed analysis of all possibilities to a future publication [33], we focus now on two of the materials with a maximum in ρ_c: Sr_2RuO_4 and κ-(BEDT-TTF)$_2$Cu(SCN)$_2$. We extract σ_c^B from the low-temperature [between 10 and 50 K c-axis resistivity of Sr_2RuO_4 and between 40 and 75 K for κ-(BEDT-TTF)$_2$Cu(SCN)$_2$] and extrapolate it to

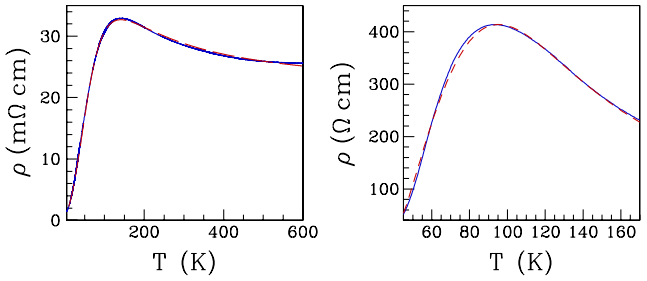

FIG. 1 (color online). ρ_c vs temperature. Solid line: experimental data; dashed line: fit into the phonon-assisted tunneling model. Left: Sr_2RuO_4 (Ref. [5]). Fitting parameters: $\sigma_{el} = 47 \times 10^3\ \Omega^{-1}\,cm^{-1}$, $\omega_D = 57$ K, and $\lambda = 18.5$. Right: κ-(BEDT-TTF)$_2$-Cu(SCN)$_2$ (Ref. [14]). Fitting parameters: $\sigma_{el} = 1.5\ \Omega^{-1}\,cm^{-1}$, $\omega_D = 140$ K, $\lambda = 18.9$.

higher temperatures. The resonant part of the conductivity is calculated numerically using Eq. (10). The fits to the data for Sr_2RuO_4 and κ-(BEDT-TTF)$_2$Cu(SCN)$_2$ are shown in the left (right) panels of Fig. 1. The quality of the fits and reasonable values of the parameters suggest that the phonon-assisted model is a viable mechanism of the c-axis anomaly at least in these compounds.

To conclude, we have shown that the Boltzmann equation and its consequences are as robust for anisotropic metals as they are for isotropic ones. The only condition controlling the validity of the Boltzmann equation is the large value of $E_F\tau$. Interplane localized states change the c-axis transport radically. An interplay between phonon-assisted tunneling and conventional momentum relaxation causes insulating or nonmonotonic dependence of ρ_c on temperature. This model is in a good agreement with the experimental data on Sr_2RuO_4 and κ-(BEDT-TTF)$_2$-Cu(SCN)$_2$.

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