Conductivity of Layered Crystals

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We show that the resistive anisotropy of an anisotropic medium is determined by the ratio of the phase coherence lengths. In layered crystals in which the interlayer transport is incoherent, the out-of-plane phase coherence length is fixed and temperature independent. This leads to a temperature dependent resistive anisotropy and to the coexistence of metallic in-plane and non-metallic out-of-plane conductivities. Our approach provides a description of the c-axis conductivity in the highly nonclassical regime, characteristic of layered cuprates.

The normal state of highly anisotropic crystals such as high-$T_c$ cuprates exhibits a number of unusual features. The coexistence of metallic in-plane ($\rho_a$) and non-metallic out-of-plane ($\rho_c$) resistivities within a range of doping is one. A crossover from metallic to non-metallic temperature ($T$) dependence is another. From a theoretical point of view, there is a great deal of interest in exploring the consequences of the idea that interlayer transport in these crystals is incoherent, so that successive interlayer transitions are uncorrelated as evidenced by the analysis of dc and infrared conductivities.

The coherence of the wave function of the charge carriers can be described in terms of the phase coherence length, hereafter called the Thouless length (TL), the distance electrons travel between dephasing inelastic collisions. An anisotropic medium, naturally, is characterized by an anisotropic TL. Below we show that, in general, the anisotropy $\sigma_a/\sigma_c$ is determined by the ratio of TL in the respective directions. The incoherence of the out-of-plane transport in layered crystals implies that TL in the c-direction is temperature independent, equal to the spacing between neighboring layers. As a result, the temperature dependence of $\rho_c/\rho_a$ is completely determined by the T-dependence of the in-plane TL. This can lead to coexistence of metallic $\rho_a$ and non-metallic $\rho_c$. We also explore the consequences of the idea that the evolution of the conductivities ($\sigma_a$ and $\sigma_c$) with temperature and doping can be described through their dependence on the in-plane Thouless length. One of the most interesting conclusions that can be drawn from this analysis is that there may exist a unifying description of the conductivity of layered crystals at different doping levels.

Let us consider an anisotropic medium characterized by three different TL: $\ell_x$, $\ell_y$, and $\ell_z$. The phase-coherent volume of such a medium, which may be considered as a block with sizes equal to the respective TL (hyperblock) has isotropic conductance; i.e., $g_x = g_y = g_z \equiv g$. One way to prove this is by using the original Thouless idea that the conductance of a block is

$$g_i = \frac{e^2}{h} \frac{dN}{dE} (\Delta E)_i,$$  

where $i = \{x, y, z\}$, $dN/dE$ is the total number of states inside the block per unit energy, and $(\Delta E)_i$ is the mean fluctuation in energy levels caused by replacing periodic by antiperiodic boundary conditions in the direction of the current. The value of $(\Delta E)_i$ is determined by the sensitivity of the energy levels to the boundary conditions, which in turn depends on the phase memory. Since the decoherence of the wavefunction occurs within the boundaries of the hyperblock, $(\Delta E)_i$ should be isotropic. Also, $(\Delta E)_i$ is determined by the time it takes for a particle to cross the system in a given direction. By definition, it takes an electron the same dephasing time $\tau_x$ to cross the hyperblock in each direction, so that $\tau_x = h/\tau_x$. A macroscopic block $\{L_x, L_y, L_z\}$ obtained by fitting together $N^3$ hyperblocks $(L_x/\ell_x = L_y/\ell_y = L_z/\ell_z = N \gg 1)$ also has isotropic conductance $G \approx Ng$ which can be expressed in terms of the components of the conductivity tensor: $G = \sigma_x L_y L_z/L_x = \sigma_y L_x L_z/L_y = \sigma_z L_y L_x/L_z$. As a result, we arrive to the following relationship between conductivities:

$$\frac{\sigma_x}{\sigma_y} = \frac{\ell_y^2}{\ell_x^2}; \quad \frac{\sigma_x}{\sigma_z} = \frac{\ell_z^2}{\ell_x^2}. \quad (2)$$

This is a general result, valid for all media, not only for layered crystals. For example, in an anisotropic Fermi liquid $\ell_i \propto V_{F,i} \tau_x$, where $V_{F,i}$ is a component of the Fermi velocity. Then, Eq. (2) reduces to the ratio of the effective masses. In the case of anisotropic diffusion, the Thouless lengths $\ell_i^2 \propto D_i \tau_x$, where $D_i$ is a component of the diffusion tensor. For such a system, Eq. (2) is equivalent to another known result $\sigma_i/\sigma_j = D_i/D_j$. Finally, for the case of variable range hopping (VRH), Eq. (2) was obtained through a different and less general approach, where $\ell_i$ is the average hopping distance in the respective direction.

We apply Eq. (2) to highly anisotropic layered crystals in which electrons lose coherence in the c-direction over the smallest possible distance, the interlayer spacing $\ell_0$ (unidirectional Ioffe-Regel limit, $\ell_x = const = \ell_0$). For simplicity, the planes are considered to be isotropic; i.e., $\ell_x = \ell_y \equiv \ell$. Under these conditions, Eq. (2) gives
\[ \sigma_c = \frac{\sigma_a \ell_0^2}{\ell^2}. \]  
(3)

Thus, the out-of-plane conductivity, which has been one of the most enigmatic features of the normal state of cuprates, is completely determined by the in-plane conductivity and the in-plane Thouless length.

Typically, quantum phenomena (e.g., superfluidity and superconductivity) reveal themselves macroscopically through phase coherence established over macroscopic distances. Here, anomalously strong interlayer decoherence in the normal state (of still unknown origin) has macroscopic consequences such as a strongly T-dependent resistive anisotropy which reflects the T-dependence of the in-plane phase coherence length. Equation (3) cannot be obtained from the quasiclassical kinetic equation because c-axis transport is highly nonclassical even when the in-plane transport can be, in principle, treated quasiclassically.

As a first example of the applicability of Eq. (3) to layered crystals, we show below that for optimally and nearly optimally doped cuprates, Eq. (3) along with diffusive in-plane transport gives a description of the out-of-plane resistivity which agrees very well with experiment and explains the observed correlation between \( \rho_a \) and \( \rho_c \). While Eqs. (2) and (3) are fundamental, the T-dependence of the in-plane TL is not, since TL can be determined by a variety of dephasing processes with one of them dominant in a certain T range for a given system. Here we assume that the strongest dephasing process is due to the thermal spread of energies, so that TL is determined by the thermal diffusion length. [Support for this assumption is provided by Ref. 11 which shows that the electron-electron interaction may be equivalent to the interaction of the electrons with the thermal fluctuations of the electromagnetic waves.] Then, the in-plane TL may be written as

\[ \ell^2 = \xi^2 + \frac{\hbar D}{k_B T}. \]  
(4)

Here we have added the empirical cutoff length \( \xi \) to the conventional definition because in a crystal, even at high temperatures, the phase coherence length cannot be shorter than a certain finite length, e.g. interatomic distance. Since elastic collisions do not lead to the loss of coherence, more realistically \( \xi \) is comparable to the elastic mean free path [at least far enough from the metal-insulator transition (MIT) as we discuss later].

With the phenomenological expression for the in-plane resistivity, \( \rho_a = \beta_a + \alpha_a T \) \[1\], Eqs. (3) and (4) give:

\[ \rho_c = \beta_c + \alpha_c T + \frac{\gamma_c}{T}, \]  
(5)

where

\[ \beta_c = \alpha_a T_0 + \beta_a \frac{\xi^2}{\ell_0^2}, \quad \alpha_c = \alpha_a \frac{\xi^2}{\ell_0^2}, \quad \gamma_c = \beta_a T_0, \]

and \( T_0 \equiv \hbar D/\ell_0^2 k_B \). Two important aspects are worth noting. First, a large constant \( \beta_c \) appears even in the absence of the intercept \( \beta_a \) (if \( \rho_a = \alpha_a T \), then \( \rho_c = \beta_c + \alpha_c T \)). This apparent "residual resistivity" violates Matthiessen's rule, because it does not scale to zero with the concentration of impurities. Second, a finite positive intercept \( \beta_a \) in \( \rho_a \) translates into a non-metallic term \( \gamma_c/T \), so that \( \rho_c \) has a minimum. Such correlations between \( \rho_a \) and \( \rho_c \) exist in optimally and slightly underdoped cuprates \[1\]. From the data of \( YBa_2Cu_3O_{7-\delta} \) \[13\] for which \( \alpha_a \approx 0.5 \mu \Omega \text{ cm}/K, \ \beta_a \approx 1.1 \mu \Omega \text{ cm}, \) and \( \alpha_c \approx 12.5 \mu \Omega \text{ cm}/K, \) we estimate \( T_0 \approx 2200 \) K, \( D \approx 4 \text{ cm}^2/\text{s}, \) and \( \xi = \lambda_0/\lambda_0^{1/2} \approx 5\lambda_0 \approx 60 \) \( \text{Å} \) (\( \lambda_0 = 11.7 \) \( \text{Å} \)). These are the least anisotropic crystals, in which a crossover from incoherent to coherent interlayer transport apparently takes place \[1\]. Nevertheless, their anisotropy is still strongly temperature dependent in agreement with Eqs. (3) and (4), and changes by a factor of 2-3 between room temperature and \( T_c \).

As a second example of the applicability of Eq. (3) to cuprates, we consider the other doping extreme - insulating crystals like \( PrBa_2Cu_3O_{7-\delta} \). The incoherence of the c-axis transport in this case means that the localized states are two-dimensional (2D). In the hopping regime, TL is given by the average hopping distance. In the VRH regime uncomplicated by Coulomb interactions, the average in-plane hopping distance \( R \) can be obtained by maximizing the hopping probability

\[ P(R) \propto \exp \left\{ -\frac{2 R^2}{A} - \frac{A}{N(R^2 - R_0^2)} \right\}, \]  
(6)

where \( \lambda \) is the localization length, \( N = \text{const} \) is the 2D density of states, and \( A \) a numerical coefficient. The only modification of the traditional treatment of VRH is the denominator \( R^2 - R_0^2 \) instead of \( R^2 \), which takes into account that two localized states with close energies cannot strongly overlap. If they do overlap, the phonon interaction that causes hopping will also hybridize them and push apart the energies of the new states. Strictly speaking, Eq. (6) is valid only for \( R \gg R_0 \sim 2\lambda \). In this limit, the average hopping distance, which in the hopping regime determines the in-plane TL, is \( \bar{R} \approx 2R_0^2/3R \), where \( \bar{R} = (\lambda A/N)^{1/3} \). Then, according to Eq. (3), \( \sigma_a/\sigma_c \approx (4R_0^2/3 + (\lambda A/N)^{2/3})/\ell_0^2 \). This T-dependence of the anisotropy \( (\alpha + bT^{-2/3}) \) was observed in insulating \( PrBa_2Cu_3O_{7-\delta} \) and superconducting \( Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta} \) \[1\]. Note that with increasing localization length \( \lambda \) (on approaching the MIT), the value of the anisotropy increases while its temperature dependence weakens \( [b/a \propto \lambda^{-4/3}] \).

In the previous two examples we considered systems far away from the metal-insulator transition. The localization length on the insulating side and the correlation
length on the metallic side of the MIT increase on approaching the transition, and therefore, at low temperatures, the phase coherence length near MIT is dominated by the divergent localization/correlation length, respectively. As a result, according to Eq. (3), at a given $\rho$, the anisotropy $\rho_c/\rho_{ab}$ changes nonmonotonically with doping, reaching a maximum in the sample closest to the MIT. Such a nonmonotonic variation of $\rho_c/\rho_{ab}$ with doping $x$ is observed in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ crystals [13]. At the same time, within a given range of temperature, the T-dependence of TL (and, hence, anisotropy) near MIT becomes substantially weaker than in either metallic [$\rho_c/\rho_a \sim 1/T$ from Eqs. (3) and (4)] or strongly insulating [$\rho_c/\rho_a \sim T^{-2/3}$] phase. This explains why the crystals of Refs. [13], which are strongly underdoped and much closer to the MIT than those of Ref. [12], exhibit a much weaker T-dependence of resistivities and anisotropy at low temperatures.

To extend the quantitative description of conductivity to all levels of doping, we use the following idea: the isotropic conductance $g$ [Eq. (1)] of the hyperblock with the sides equal to the respective TL $\{\ell, \ell, \ell_0\}$ can be described as a function of $\ell$ only, so that the temperature and magnetic field dependence of $g$ appears exclusively through that of $\ell$. The justification of this hypothesis can be partly found in the ideas of the scaling theory [14]. Indeed, it is known [14] that the conductivity of a conventional 3D Fermi liquid, as well as the quantum corrections [15], can be expressed as functions of TL with no explicit temperature dependence. For a medium "composed" of hyperblocks $\{\ell, \ell, \ell_0\}$, the in-plane conductivity $\sigma_a$ differs from conductance $g$ by a constant; i.e., $\sigma_a(\ell) = g(\ell)/\ell_0$.

Therefore, below we discuss the $\sigma_a(\ell)$ dependence instead of $g(\ell)$. This assumption that the T-dependence of the conductivities enters only through that of $\ell$, immediately explains why metallic $\sigma_a$ and nonmetallic $\sigma_c$ can coexist. For example, if $\sigma_a \propto \ell^\nu$ with $0 < \nu < 2$, then, according to Eq. (3), $\sigma_a$ is metallic ($d\sigma_a/d\ell > 0$) while $\sigma_c$ is nonmetallic ($d\sigma_c/d\ell < 0$).

A quantitative description of the $\sigma_a(\ell)$ dependence can be given through the logarithmic derivative

$$\kappa = \frac{d\ln \sigma_a}{d\ln \ell}$$

which we treat as a function of $\sigma_a$. This definition formally resembles the "trajectories" of the scaling theory [14]. The difference is that Ohm's law does not impose any restrictions on the $\sigma_a(\ell)$ dependence and the limiting values of $\kappa$. Instead, we rely on experimental data to infer the shape of the trajectory $\kappa(\sigma_a)$. We can do this because, according to Eq. (3), the measured anisotropy $\eta \equiv (\rho_c/\rho_a)^{1/2}$ gives directly the temperature dependence of TL; i.e., $\ell = \eta_0$. Therefore, the experimental dependence $\sigma_a$ vs. $\eta$ determines $\kappa(\sigma_a)$ and $\kappa(\sigma_a) = \ln \sigma_a/d\ln \eta$ dependsences. The thick segments in Fig. 1 are schematic representations of $\kappa(\sigma_a)$ for several levels of doping, where the range of $\sigma_a$ corresponds to "accessible" temperatures $T_{min} < T < T_{max}$ ($T_{max}$ is typically $300 - 350$ K and $T_{min} \approx T_c$ in superconducting crystals). The thin lines indicate the hypothetical extensions of the trajectories outside of this range. According to Eq. (3),

$$\frac{d\ln \sigma_a}{d\ln \ell} = \kappa - 2.$$

Therefore, both $\sigma_a$ and $\sigma_c$ are metallic for $\kappa > 2$ and nonmetallic for $\kappa < 0$, while metallic $\sigma_a$ and nonmetallic $\sigma_c$ coexist for $0 < \kappa < 2$.

Segment 1 in Fig. 1 represents optimally or overdoped regimes where both conductivities $\sigma_a$ and $\sigma_c$ are metallic at all temperatures $T > T_{min}$ because the whole segment is located above the threshold $\kappa = 2$. The previous example of optimally doped cuprates [$\rho_0 = \alpha_aT$, $\ell$ given by Eq. (4), and $\rho_c = \beta_c + \alpha_c T$] leads to $\sigma_a = g(\ell^2 - \xi^2)$ and $\kappa(\sigma) = 2 + \xi/\sigma$, where $q = k_B/\alpha_a hD$ and $\xi = 2q\xi^2$.

Segment 2 corresponds to a slightly underdoped system. At high temperatures (small $\sigma_a$) $\kappa > 2$ and, therefore, $\sigma_c$ is metallic. With increasing $\ell$ (decreasing $T$) $\sigma_c$ increases, reaches a maximum when $\kappa(\sigma_a) = 2$, and then decreases. Therefore, $\rho_c$ changes from metallic at high $T$ to nonmetallic at low $T$, similar to the T-dependence given by Eq. (5). Within the same range of temperature, $\rho_a$ is metallic.
Segment 3 represents a moderately underdoped system. It lies entirely within the range \(0 < \kappa < 2\) and corresponds to metallic \(\rho_a\) and nonmetallic \(\rho_c\) for all \(T_{min} < T < T_{max}\).

Segment 4 corresponds to strongly underdoped crystals \([14, 13]\) with \(\sigma_a\) changing from metallic at high \(T\) (\(\kappa > 0\)) to nonmetallic at lower \(T\) (\(\kappa < 0\)). The singularity \(\kappa = 0\) is integrable:

\[
\kappa(\sigma) \approx \pm \zeta \left( \ln \frac{\sigma_{max}}{\sigma} \right)^{1/2},
\]

so that \(\sigma_a(\ell)\) as determined by the equation

\[
\int_{\sigma_{max}}^{\sigma} \frac{d \ln \sigma}{\kappa(\sigma)} = \ln \frac{\ell}{\ell_1}
\]

reaches the maximum value \(\sigma_{max}\) at a finite \(\ell = \ell_1\) and decreases with further increasing \(\ell\) (decreasing \(T\)). Equations (7) and (8) give \(\sigma_a(\ell) \approx \sigma_{max} \exp\{-2 \zeta^2 \ln^2(\ell/\ell_1)/4\} \).

Finally, segment 5 corresponds to an insulating crystal. In this case, \(\ell \approx R\) and \(\sigma_a \sim \sigma_0 \exp\{-3\ell/\lambda\}\). Therefore, \(\kappa(\sigma) \approx \ln(\sigma/\sigma_0)\).

As shown, all five curves in Fig. 1 represent the same \(\kappa(\sigma/\bar{\sigma})\) dependence, shifted with respect to each other due to different values of \(\bar{\sigma}\) (determined by the density of carriers \(n\) and other parameters which cannot be absorbed into TL). The reduction of \(n\) by doping reduces \(\bar{\sigma}\), shifting the respective segment of \(\kappa(\sigma/\bar{\sigma})\) to lower absolute values of \(\sigma\) and, at the same time, ahead along the trajectory in terms of the reduced variable \(\sigma/\bar{\sigma}\). Integration similar to Eq. (8) gives \(\sigma(\ell)\) of the form

\[
\frac{\sigma_a}{\sigma} = f \left( \frac{\ell}{\ell} \right),
\]

where \(f(y)\) is a universal function \([f(1) = 1]\), so that the conductivity of the crystals at different levels of doping is described by the same \(f(\ell)\) dependence, provided the appropriate choice of doping dependent parameters \(\bar{\sigma}\) and \(\ell\). This hypothesis can be verified experimentally by checking whether the conductivity at different doping levels plotted against anisotropy \(\eta\) form a continuous curve when \(\sigma_a\) and \(\eta\) are properly normalized.

This idea of universality of the \(\sigma_a(\ell)\) dependence allows the prediction of the behavior of the normal state conductivities at temperatures below \(T_c\). For example, let segment 3 in Fig. 1 represent a superconducting crystal with metallic \(\sigma_a\) and nonmetallic \(\sigma_c\) for all \(T_c < T < T_{max}\). If we suppress the onset of superconductivity with a magnetic field in order to reveal the normal state at \(T < T_c\), we will observe that the trajectory crosses the threshold \(\kappa = 0\) and \(\sigma_a\) also becomes nonmetallic. This type of development has been observed \([13, 14]\) in underdoped cuprates.

It is possible that the trajectories shown in Fig. 1 oversimplify the situation, and there may be more than one type of \(f(\ell)\) dependence. The data suggest that in \(Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}\), a metal-insulator transition takes place at intermediate doping levels somewhere between optimally doped and strongly insulating phases \([13]\). As a result, there may be at least two types of trajectories (or a branching point), corresponding to the phases on either side of the MIT.

The assumption that the response of the conductivities to external perturbations other than temperature also comes from that of TL allows us to predict these responses because

\[
\frac{\partial \ln \sigma_a}{\partial x} = \kappa \frac{\partial \ln \ell}{\partial x}; \quad \frac{\partial \ln \sigma_c}{\partial x} = (\kappa - 2) \frac{\partial \ln \ell}{\partial x}.
\]

Here \(x\) can be magnetic field or concentration of impurities. For \(\kappa > 2\) or \(\kappa < 0\) both conductivities change similarly. However, for a wide range of doping levels and temperatures \([\text{Fig. 1}]\), for which \(0 < \kappa < 2\), the two conductivities respond oppositely.

For example, in \(Bi_2Sr_2CaCu_2O_8+y\) single crystals \(\sigma_c\) increases and \(\sigma_a\) decreases with increasing \(Zn\) concentration \([13]\). The impurities reduce the in-plane TL (\(\partial \ell/\partial x < 0\)) by decreasing the elastic mean free path and the diffusion coefficient. The crystals used \([13]\) exhibit metallic \(\rho_a(\ell)\) and nonmetallic \(\rho_c(\ell)\), so that \(0 < \kappa < 2\) and, as a result, \(\partial \sigma_a/\partial x < 0\) while \(\partial \sigma_c/\partial x > 0\). Sometimes, the increase of the c-axis conductivity in response to a perturbation has been interpreted in literature as a crossover to coherent transport in the c-direction. We see that this is not necessarily the case. The anisotropy may decrease and \(\sigma_c\) increase due to the reduction of the in-plane TL, even when the c-axis TL remains fixed.

The effect of the magnetic field \(H\) on conductivities is determined by the destructive interference \([3]\), resulting in the reduction of TL (\(\partial \ell/\partial H < 0\)). Therefore, according to Eq. (3), the magnetoanisotropy \(\bar{\delta}(\rho_c/\rho_a)\) should be negative, while the sign and relative magnitude of the magnetoconductivities \(\Delta \sigma_a\) and \(\Delta \sigma_c\) depend on the value of \(\kappa\) according to Eq. (10). One can see that the magnetoconductivities correlate with the temperature coefficient of the respective component of the resistivity:

\[
\frac{\partial \rho_{a,c}}{\partial H} = Q \frac{\partial \rho_{a,c}}{\partial T},
\]

where \(Q = (\partial \ell/\partial H)/(\partial \ell/\partial T)\). Since \(Q > 0\), the sign of magnetoresistance \(\Delta \rho_{a,c}/\partial T\) is the same as the sign of \(\partial \rho_{a,c}/\partial T\), as indeed was reported in Refs. \([3, 12, 13]\). The magnitude of the magnetoeffects depends on the number of flux quanta \((h_i)\) permeating the hyperblock. For \(H || c, h_c = H \ell^2/\phi_0 = (\rho_c/\rho_a) H/\ell_0 (H_0 = \ell_0/\phi_0, \phi_0\) is the quantum of flux). If the field is parallel to the planes, \(h_a = H \ell_0/\phi_0 = (\rho_a/\rho_c)^{1/2} H/\ell_0\). Thus, \(h_c \gg h_a\) which may result in a strong angular dependence of the magnetoeffects.
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