Indications of coherence-incoherence crossover in layered transport

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For many layered metals the temperature dependence of the interlayer resistance has a different behavior than the intralayer resistance. In order to better understand interlayer transport we consider a concrete model which exhibits this behavior. A small polaron model is used to illustrate how the interlayer transport is related to the coherence of quasi-particles within the layers. Explicit results are given for the electron spectral function, interlayer optical conductivity and the interlayer magnetoresistance. All these quantities have two contributions: one coherent (dominant at low temperatures) and one incoherent (dominant at high temperatures).

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Many of the most interesting strongly correlated electron materials have a layered crystal structure and highly anisotropic electronic properties. Examples include the cuprates \^[1, 2, 3\], colossal magneto-resistance materials \^[4, 5\], organic molecular crystals \^[6, 7\], and strontium ruthenate \^[8\]. One particularly poorly understood property of these materials is that often the resistivity perpendicular to the layers has quite a distinct temperature dependence to that parallel to the layers \^[9\]. This is in contrast to what is expected for an anisotropic Fermi liquid: the parallel and perpendicular resistivity then have the same temperature dependence, being determined by the intralayer scattering rate, \(\Gamma(T)\). (This result holds even when \(\Gamma(T)\) is larger than the interlayer bandwidth \^[10\]).

In many of these materials the interlayer resistivity is a non-monotonic function of temperature with a maximum at some temperature \(T_{\perp}^{\max}\). In some of the materials the interlayer resistivity also has a maximum as a function of temperature, but at a higher temperature \(T_{\parallel}^{\max} > T_{\perp}^{\max}\). Previously it has been suggested that the maximum in the temperature dependence of the interlayer resistivity is associated with a crossover from coherent interlayer transport at low temperatures to incoherent transport at high temperatures. There is no consensus as to what actually determines \(T_{\perp}^{\max}\). Recent angle resolved photoemission spectroscopy (ARPES) experiments on two different layered cobalt oxide compounds \^[11\] found that one only observed peaks in the electronic spectral function (corresponding to coherent quasi-particle excitations within the layers) below a temperature \(T_{\text{coh}}\) that was comparable to \(T_{\perp}^{\max}\).

Although many theoretical papers have considered the problem of incoherent interlayer transport (see for example Ref. \^[12\] and the references given in Ref. \^[11\]), we are unaware on any theory which starts with a many-body Hamiltonian and produces the three temperature scales, \(T_{\text{coh}}, T_{\perp}^{\max}, \text{and } T_{\parallel}^{\max}\). In this paper we consider a simple microscopic model which exhibits \(T_{\text{coh}} \sim T_{\perp}^{\max} < T_{\parallel}^{\max}\). The model is a layered version of Holstein’s molecular crystal model where the electrons strongly couple to bosonic excitations to produce small polarons. It should be stressed that we are not claiming that the charge transport involves small polarons in all of the above materials. Rather, we suggest that this model can provide insight into the relevant physics associated with these temperature scales.

We start with a Holstein model \^[13\] for an infinite layered system where the electrons interact with dispersionless bosons. The Hamiltonian is

\[
H = \sum_i \epsilon^0 c_i^\dagger c_i + \hbar \omega_0 \sum_i a_i^\dagger a_i + \sum_{\langle i\eta\rangle} t_{i\eta} c_i^\dagger c_{\eta},
\]

\[
+ M \sum_i c_i^\dagger c_i (a_i + a_i^\dagger),
\]

where \(\epsilon^0\) is the on-site energy, \(\omega_0\) is the characteristic frequency of the bosons, \(t_{i\eta}\) is the hopping integral between nearest-neighbor sites \(i\) and \(\eta\), \(M\) is the coupling between the bosons and the electrons. We introduce a dimensionless coupling \(g = \left(\frac{M}{\omega_0}\right)^2\). We have that \(g \gg 1\).

The nature of the transport depends on how \(t_{\parallel}\) and \(t_{\perp}\) compares with \(\Gamma\), the scattering rate due to the bosons. We assume that that \(\Gamma \gg t_{\perp}\), so that the inter-layer transport can be described by considering two decoupled layers. The Hamiltonian can be specified for this system. Two layers are coupled with a hopping Hamiltonian. Within each layer the electrons can hop but there is a coupling to a bosonic degree of freedom in each layer with characteristic frequency \(\omega_\eta\). The bosons can be phonons, magnons, plasmons, or particle-hole excitations. We only consider a single frequency \(\omega_\eta\) for reasons of simplicity; it allows us to express some of our results in a analytical form.

We then use the Hamiltonian:

\[
H = H_1 + H_2 + H_4
\]
where
\[ \mathcal{H}_1 = \sum_i \epsilon_i c_i^\dagger c_i + \hbar \omega_0 \sum_i a_i^\dagger a_i + t_{\parallel} \sum_{<ij>} c_i^\dagger c_j \]
\[ + M \sum_i c_i^\dagger (a_i + a_i^\dagger), \]
\[ \mathcal{H}_2 = \sum_j \epsilon_j d_j^\dagger d_j + \hbar \omega_0 \sum_j b_j^\dagger b_j + t_{\parallel} \sum_{<ij>} d_i^\dagger d_j \]
\[ + M \sum_j d_j^\dagger d_j (b_j + b_j^\dagger), \]
\[ \mathcal{H}_t = t_{\perp} \sum_i (c_i^\dagger d_i + \text{h.c.).} \]

Here, and below, \( c, i, a, 1 \) refers to one layer, and \( d, j, b, 2 \) to the other one. First we focus on the properties of the two separate layers. We perform a Lang-Firsov transformation [13] to remove the coupling of the electrons to the bosons. Then \( c_i \rightarrow \tilde{c}_i = c_i X_i \) where
\[ X_i = \exp \left[ \frac{M}{\hbar \omega_0} (a_i - a_i^\dagger) \right], \]
and \( a_i \rightarrow a_i \frac{M}{\hbar \omega_0} c_i \). The Hamiltonian is transformed to
\[ \tilde{\mathcal{H}} = e^S \mathcal{H} e^{-S} \] where \( S = \frac{M}{\hbar \omega_0} \sum_i \tilde{c}_i^\dagger \tilde{c}_i \). This diagonalizes the electron-boson part of the Hamiltonian, but introduces extra \( X \)-operators [14] in the hopping parts of the Hamiltonian. For example:
\[ \tilde{\mathcal{H}}_t = t_{\perp} \sum_{<ij>} (c_i^\dagger X_i^\dagger d_j Y_j + \text{h.c.),} \]

The intralayer hopping terms can be treated by adding and subtracting to the Hamiltonian a term
\[ t_{\parallel} \sum_{<ij>} \langle X_i X_j^\dagger \rangle c_i^\dagger c_j = \sum_k \epsilon_k c_k^\dagger c_k \]
where \( \langle . . \rangle \) denotes a thermal average over boson states and this term describes a tight-binding band of small polarons within each layer [14] [16].
\[ \epsilon_k = \epsilon^0 - e^{-g(1+2n_B) t_{\parallel} \cos(k_x a) + \cos(k_y a)}, \]
where \( a \) is the lattice constant within the layers and \( n_B(T) = (\exp(\hbar \omega_0/k_B T) - 1)^{-1} \) is the Bose function.

There is then a residual interaction [16] between the polarons and the bosons which is described by
\[ \tilde{\mathcal{H}}_{p-b} = t_{\parallel} \sum_{<ij>} (X_i X_j^\dagger - \langle X_i X_j^\dagger \rangle) c_i^\dagger c_j, \]
and leads to scattering of the small polarons. The first non-zero contribution to the imaginary part of the polaron self-energy, \( \Sigma \), comes when the polaron emits one boson and absorbs one boson. If the energy dependence of the the density of states (DOS) is neglected one finds that [14].
\[ \Gamma(T) = W g^2 n_B(T)(1 + n_B(T)), \]
where \( W \) is the renormalized bandwidth, \( W = 4 t_{\parallel} e^{-g(1+2n_B)} \) [14] [15] [16].

Note that the small polarons are composite particles (quasi-particles). They consist of an electron bound to a "cloud" of bosons. This coherent quantum state can move freely within the layers producing coherent charge transport. In contrast, ARPES involves ejection of electrons rather than polarons from the crystal. Similarly, the interlayer charge transport involves the tunneling of electrons between layers. In order for this to occur the bosons bound to the electron in the polaron must be removed, the electron tunnels, and a new set of bosons are bound to the electron.

\textbf{Greens function within a single layer.} Let us start by calculating the Matsubara Green function (GF) within one layer, ignoring the coupling between the layers. After the Lang-Firsov transformation we calculate the small polaron GF
\[ G^0(\mathbf{k}, \tau) = -i \Theta(\tau) \frac{1}{N} \sum_{i,i'} e^{i \mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_{i'})} \langle T_\tau \tilde{c}_{i'}(\tau) \tilde{c}_i(0) \rangle \]
\[ = -i \Theta(\tau) e^{i \epsilon_{\mathbf{k}} \tau / \hbar}, \]
where \( \Theta(\tau) \) is the step function, \( T_\tau \) is the time ordering operator, \( \mathbf{R}_i \) is the position of lattice site \( i \). \( \Gamma \) is the inverse lifetime associated with the interaction, Eq. (3), and there are \( N \) lattice sites in each layer.

The electron GF, \( G(\mathbf{k}, i \omega_n) \), involves a convolution of the polaron GF with the Fourier transformed (denoted \( \mathcal{F} \)) \( X \)-operators [17].
\[ G(\mathbf{k}, i \omega_n) = \frac{1}{N} \sum_{\omega_n, i, i', k'} \mathcal{F}\{ \langle X_{i'}(\tau) X_i^\dagger(0) \rangle \} G^0(\mathbf{k}', i \omega_n') e^{i (\mathbf{k} - \mathbf{k}') \cdot (\mathbf{R}_{i'} - \mathbf{R}_i)} \]
Performing the summation over \( \mathbf{R}_{i'} - \mathbf{R}_i \) we obtain
\[ G(\mathbf{k}, i \omega_n) = e^{-g(1+2n_B) \left\{ \frac{1}{i \omega_n - \epsilon_k + i \Gamma} \right\}} + \sum_{k'} I_{0} \frac{2 g \sqrt{n_B(1 + n_B)}}{i \omega_n - \epsilon_{k'} + i \Gamma} - 1 \]
\[ + \sum_{k', l \neq 0} I_{l} \frac{2 g \sqrt{n_B(1 + n_B)}}{i \omega_n - \epsilon_{k'} + i \hbar \omega_0 / 2} \}
\]

\textbf{FIG. 1:} The two layers with polarons are coupled by a hopping Hamiltonian.
The quasi-particle peak which occurs for the boson modes become populated, for different temperatures. With increasing temperature \( f \) Dirac distribution function \( g \) boson coupling of \( \delta \) increases and \( T/h-B \) at higher temperature. The results are shown for an electron - A Ranninger [17].

The product of the spectral function \( A(\mathbf{k}, \epsilon) \) with the Fermi-Dirac distribution function \( f(\epsilon) \) is shown because this can be compared with ARPES spectra. Note that the well-defined quasi-particle peak which occurs for \( k_B T \ll \hbar \omega_0 \) disappears at higher temperature. The results are shown for an electron-boson coupling of \( g = 1 \). The inset shows the same thing for a smaller bandwidth.

Note that the GF is a sum of a coherent and an incoherent part, i.e., the terms on the second and third lines are independent of \( \mathbf{k} \). Eq.(7) is a generalization to non-zero temperatures of the GF found by Alexandrov and Ranninger [17].

In Fig. 2 we plot the electron spectral function, \( A(\mathbf{k}, \omega) = \text{Im}[G(\mathbf{k}, \omega)] \), resulting from the GF, Eq.(7), for different temperatures. With increasing temperature the boson modes become populated, \( n_B(T) \) increases and the spectral weight shifts from the coherent part of the spectral function to the incoherent part.

Qualitatively similar behavior was seen in recent ARPES [11] measurements. This behavior does not change much qualitatively when \( g \) is changed. From plots we estimated that the crossover takes place at

\[
k_B T_{\text{coth}} \sim \frac{\hbar \omega_0}{2g}.
\]

This can also be justified using Eq.(7) when \( W < \hbar \omega_0 \).

**Interlayer conductivity.** Let us derive an expression for the current perpendicular to the planes. We assume that the hopping between the layers only takes place between nearest neighbors (see Fig. 3). At applied field \( V \) the current is given by

\[
I_{\mu\nu}(eV) = \frac{2e}{h} \text{Im} \left[ \int_0^\beta d\tau e^{i\epsilon V} \tau \langle \hat{T}_\tau \hat{j}_\mu(\tau) \hat{j}_\nu^\dagger(0) \rangle \right],
\]

where \( \hat{j} \) is the current operator, \( \mu \) and \( \nu \) are directions in the crystal.

Using the Hamiltonian above and Eq.(9) we have, to second order in \( t_\perp \),

\[
I_{\perp}(eV) = \frac{2e}{h} t_\perp^2 \sum_{j,j_1} \int_0^\beta d\tau e^{i\epsilon V} \tau \times \left( \left[ T_\tau e^{j_1}(\tau)d_j^\dagger(\tau)c_{j_1}(0) \right] - \left[ T_\tau Y_j^\dagger(\tau)Y_j(\tau)X_{j_1}^\dagger(0)X_{j_1}(0) \right] \right),
\]

where \( d \) is the distance between the 2 layers.

We assume that the bosons in separate layers are independent of one another. Hence, in Eq.(10), we can decouple the \( X \) and \( Y \) polaron operators corresponding to the first and second layers. This means that the Fourier transformed averages of the electron operators gives rise to two GFs. These GFs describe polarons bands within each layer.

After some algebra we can write the current as

\[
I_{\perp}(eV) = \frac{2e}{h} t_\perp^2 \sum_{j,j_1} \int_0^\beta d\tau e^{i\epsilon V} \tau \times \left( \left[ T_\tau e^{j_1}(\tau)d_j^\dagger(\tau)c_{j_1}(0) \right] - \left[ T_\tau Y_j^\dagger(\tau)Y_j(\tau)X_{j_1}^\dagger(0)X_{j_1}(0) \right] \right),
\]

where \( \mathbf{p} \) belongs to the first (1) layer and \( \mathbf{k} \) to the second (2) layer. \( A^0_1 \) and \( A^0_2 \) are the spectral functions for the polaron GFs in each layer respectively, and \( I_l \) is a modified Bessel function of order \( l \).
Note the similarity in structure between Eq.(11) and Eq.(10). The expression for the current, Eq.(10), has a contribution from coherent and one from incoherent transport. The first line corresponds to tunneling where the momentum of the polaron parallel to the layers is conserved. In the second and third terms the intralayer momentum is not conserved. The third line has a difference energy of $\hbar \omega_0$ between the polarons in the two layers because there is a non-zero difference between the net number of bosons that are absorbed and emitted in the tunneling event. At low temperature the coherent part dominates but at high temperature ($k_B T > \hbar \omega_0$) the incoherent mechanism of transport will dominate.

Thus, there is a crossover from coherent to incoherent transport.

Then, the inter-layer conductivity is easily obtained 

$$\sigma_\perp = e^2 \frac{dI_0}{d(\epsilon V)} \bigg|_{\epsilon V = 0} \frac{d\epsilon}{dV}.$$  

This should be multiplied with the number of sites in one layer. The interlayer resistivity, measured in experiments, is simply $\sigma_\perp^{-1}$.

By using the fact that when $\Gamma \ll W$, $\sum_{k} A(k, \epsilon) = D(\epsilon)$, where $D(\epsilon)$ is the DOS, and assuming that terms containing the derivative of the spectral function is small $dA(\epsilon + eV)/dV \ll A(\epsilon + eV)D(\epsilon)$ we can write the conductivity as:

$$\sigma_\perp = \frac{2e^2}{\hbar} t_\perp^2 \int d\epsilon \sum_{k} A^2(k, \epsilon) \left[ -\frac{d\epsilon}{d\epsilon} \right], \quad (12)$$

where $A(k, \epsilon)$ is the electron spectral function for a single layer. Directly substituting Eq.(6) in this we can obtain Eq.(11).

We can estimate the temperature of the crossover in the conductivity by having equal contribution from the coherent and the incoherent parts. If we look at the conductivity there is a minima (maxima in the resistivity), corresponding to the crossover (see inset in Fig.3). Ignoring the contribution from the $l \neq 0$ terms in Eq.(11) we can get an approximate expression for the crossover temperature for the interlayer resistivity:

$$k_B T_\perp^{\text{max}} \sim \frac{\hbar \omega_0}{\sqrt{2}g} \sim 0.59 \frac{\hbar \omega_0}{g}. \quad (13)$$

This expression compares quite well to the crossover temperature extracted from a numerical plot of the resistivity versus temperature in Fig.3. Hence, we see that $T_\parallel^{\text{coh}}$ and $T_\perp^{\text{max}}$ are comparable. At these temperatures, $\Gamma \sim 0.3 W$ A more detailed analysis shows that the $l \neq 0$ terms in Eq.(11) contributes linearly in $W$. So the estimate for the crossover, Eq.(13), should only hold when $W \lesssim \hbar \omega_0$, but extensive numerical work showed that it has a wide range of validity.

In general, the interlayer conductivity for decoupled layers is $\sigma_{\text{ll}}$,

$$\sigma_\perp = \frac{2e^2}{\hbar} t_\perp^2 \int d\epsilon \sum_{k} A^2(k, \epsilon) \left[ -\frac{d\epsilon}{d\epsilon} \right], \quad (12)$$

where $A(k, \epsilon)$ is the electron spectral function for a single layer. Directly substituting Eq.(6) in this we can obtain Eq.(11).

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It is interesting to note that $\sigma^{\text{coh}} \sim \sigma^{\text{incoh}}$ actually occurs at a temperature lower than $T_\perp^{\text{max}}$, as can be seen in the inset in Fig.3. This is because an almost constant contribution from the $l \neq 0$ terms.

**Intralayer conductivity.** When we calculated the current between the layers we assumed that the small polaron band was well-defined. However, it is known that even within the layer, as the temperature increases, there is a crossover from coherent to incoherent transport, occurring at $T_\parallel^{\text{max}}$, and there is a maximum in the resistivity associated with this crossover [13]. An important question is the size of $T_\parallel^{\text{max}}$ relative to $T_\perp^{\text{max}}$.

We are unaware of a systematic method of calculating the full temperature dependence of the intralayer conductivity. At low temperatures it can be calculated by considering the band transport of local polarons. At high temperatures one can consider the diffusion (or hopping) of localized polarons. Extrapolating the results to intermediate temperatures provides a means to estimate the crossover temperature and the regions of validity of the different expressions.

At low temperatures ($\Gamma \ll W$) the transport in the layers is coherent. We assume that we have well developed quasi-particles in the layers and the bosons acts as a small perturbation. The conductivity is given by:

$$\sigma_\parallel = \frac{e^2}{2\pi^2} \int d\epsilon \int d^2k \frac{\epsilon_k^2}{\Gamma} \delta(\epsilon - \epsilon_k) \left( -\frac{d\epsilon}{d\epsilon} \right).$$

We use a tight binding model for the electrons to calcu-
late the velocities, and the decay $\Gamma$ is taken from Eq. (9) above.

At high temperatures ($\Gamma \gg W$) the electron is trapped by the formation of the polaron. The electrons are localized at the lattice sites and the concept of a wave-vector for the small polaron is meaningless. The intralayer hopping term in the Hamiltonian should then be treated as the perturbation. The intralayer conductivity at high temperatures from Eq. (8) depends on the polaron GF, solved when the hopping term is the perturbation. However, due to the lack of dispersion of the bosons the time integral in the conductivity diverges [15]. We overcome this problem by including a finite self energy in the electron GF it has a self energy that satisfies

$$\Sigma(\omega) = 4\hbar^2 G \frac{1 + r^2 G^2 + r^4 G^4}{(1 - t^2 G^2)^3}. \quad (14)$$

This is a self-consistent equation for the real and imaginary part of the self energy that have to be solved simultaneously to self consistency. This was done on a square lattice where the polaron can hop to four different sites each time. The conductivity for the low and high temperature regions were plotted and the maxima in $\rho_\parallel$ was extracted.

There is a crossover also for the intralayer resistivity. From Fig. 3 the crossover occurs roughly when

$$k_B T_{\parallel}^{\max} \sim 2 \frac{\hbar\omega_0}{g}, \quad (15)$$

using this in Eq. (8), means that the crossover occurs when $\Gamma \sim 8W$. The intralayer crossover occurs at higher temperatures than the interlayer crossover, therefore the assumption made above that for the interlayer calculation we have well developed quasi-particles within each layer is justified.

**Optical conductivity.** The frequency dependence of the interlayer optical conductivity, $\sigma_\perp(\omega)$, has been suggested to be a probe of interlayer coherence in the metallic state [18]. The optical conductivity can be found from Eq. (8), by letting $eV \rightarrow \omega$, and calculating the derivative.

Fig. 4 shows how at low temperatures there is a well-defined Drude peak at zero frequency due to coherent interlayer transport of small polarons. The width of this feature is approximately $\Gamma$. Note that this feature occurs even though $\Gamma > t_\perp$, as has been pointed out previously [19]. As the temperature increases the spectral weight of this feature decreases and is replaced with a broader feature associated with incoherent interlayer transport and with a width that is determined by the small polaron bandwidth within the layers. The incoherent part becomes narrower with increasing temperature because of the polaron narrowing of the bands. Changing $g$ and $t_\parallel$ does not qualitatively change this temperature-dependent crossover.

**Magnetoresistance.** If we apply a magnetic field, $B$, parallel to the layers (the x-y plane) we have an orbital effect on the paths of the electrons. This can be described by a shift in the Bloch wave vector, $k \rightarrow k - \frac{e}{\hbar}A$, where $A$ is the vector potential for the magnetic field. For a magnetic field in the x direction, when an elec-
tron tunnels between adjacent layers it undergoes a shift in the y-component of its wave vector by \(-dB\). In the general expression Eq.\((12)\), \(A^2(k, \epsilon)\) is replaced with \(A(k, \epsilon)A(k + \frac{\epsilon}{T}dB\hat{y})\). However, since the incoherent part of the conductivity contains a summation over \(k\)-space and is independent of \(k\), this will be unaffected by the magnetic field. Thus, we will have two contributions to the interlayer conductivity and one is \(B\)-independent:

\[
\sigma_\perp(B) = \sigma_{\perp, \text{coh}}(B) + \sigma_{\perp, \text{incoh}}(B = 0),
\]

\(\sigma_{\perp, \text{coh}}(B)\) decreases with increasing magnetic field \([14, 20]\).

\[
\sigma_{\perp, \text{coh}}(B) = \frac{\sigma_{\perp, \text{coh}}(B = 0)}{\sqrt{1 + (evFcBt)^2}}
\]

where \(v_F\) is the Fermi velocity. If we increase \(B\), the coherent part decreases, and, therefore, \(T_{\text{max}}^{\perp}\) would shift to lower values. A separation of the conductivity in two parts, as in Eq.\((13)\), has been proposed previously on a phenomenological basis, in order to describe the magneto-resistance of \(\text{Sr}_2\text{RuO}_4\) \([3]\). (Except there a weak field dependence is associated with the incoherent contribution due to Zeeman splitting).

We have shown that a small polaron model for transport in layered systems shows a crossover from coherent to incoherent transport at different temperatures for intralayer and interlayer transport. It can be observed in ARPES, as well as in measurements of magneto-resistance and optical conductivity. It is sometimes suggested (or assumed) that the maximum in the interlayer resistivity as a function of temperature occurs at a temperature \(T_{\text{max}}^{\perp}\) determined by the strength of the interlayer hopping \(t_{\perp}\), either by \(k_B T_{\text{max}}^{\perp} \sim t_{\perp}\) or \(\Gamma(T_{\text{max}}^{\perp}) \sim t_{\perp}\) where \(\Gamma(T)\) is the temperature dependent scattering rate within the layers. However, we find that \(T_{\text{max}}^{\perp}\) can occur at a higher temperature, which is actually independent of \(t_{\perp}\), and instead closely related to \(T_{\text{coh}}^{\perp}\).

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