CHARGE TRANSPORT IN SYNTHEtic METALS

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The phenomenology of charge transport in synthetic metals is reviewed. It is argued that the conventional quasiparticle picture and Boltzmann transport theory do not apply to these materials. The central ideas of Fermi liquid theory are reviewed, and the significant corrections produced by quasiparticle scattering from ferromagnetic spin fluctuations in liquid \textsuperscript{3}He are described. It is shown that Sr$_2$RuO$_4$ does not display the symptoms of a nearly-ferromagnetic Fermi liquid, so the source of its odd angular momentum pairing remains to be understood. The solution of an assisted-tunneling model of charge transport in quasi-one dimensional materials is described. This model has a quantum critical point and gives a resistivity that is linear in temperature or frequency, whichever is greater.

1 The Nature of the Problem

The twentieth century has seen the development of an outstandingly successful theory of the solid state, including a theory of the equilibrium and transport properties of metals.\textsuperscript{1} For transport properties, the central concept of a mean free path $l$ was introduced at the beginning of the century.\textsuperscript{2,3} However a number of interesting synthetic metals discovered in the past few decades seem to violate the conventional theory. They are “bad metals”\textsuperscript{4} in the sense that their resistivity $\rho$ has a metallic temperature dependence (it increases with the temperature $T$) but, if $\rho$ were interpreted in terms of a mean free path, the uncertainty $2\pi/l$ in the Fermi wave vector $k_F$ would be greater than $k_F$ itself, so the concept of an electron state in momentum space would be ill defined. This is known as the Ioffe-Regel condition.\textsuperscript{5} Among the materials in question are the high temperature superconductors; other oxides including the ruthenates, such as SrRuO$_3$ and Sr$_2$RuO$_4$; organic conductors, such as TTF-TCNQ; and alkali-doped C$_{60}$.

In all of these materials, $\rho$ decreases as $T$ decreases and, unless a phase transition intervenes, $\rho$ could become small enough to give the appearance that the conventional theory is applicable. However, for many synthetic metals, $\rho$ is linear in $T$ over an extended range of temperatures, and it would be inconsistent to explain one and the same phenomenon by the conventional theory at low temperatures, and by some other theory at high temperatures.

Another issue is the role of phonons. Typically the $T$-linear resistivity extends from well above to well below phonon Debye temperatures, so it cannot
be attributed to scattering from classical phonons. But there is no change in
the slope of $\rho$, so it appears that the phonons make no contribution at all to
the resistivity, even in materials in which there is independent evidence of a
significant electron-phonon coupling. This behavior is quite inconsistent with
the usual Boltzmann transport theory in which scattering rates are additive.

Alkali-doped $C_{60}$ is a particularly interesting example of a bad metal be-
cause $\rho$ is $T$-linear but, in a quasiparticle picture, it would correspond to a
mean free path that is less than the size of a $C_{60}$ molecule, and can even be
as small as a carbon-carbon distance which is less than 10% of the size of a
unit cell. Then it clearly does not make sense to interpret transport in terms
of quasiparticles freely propagating from cell to cell.

TTF-TCNQ is an organic conductor consisting of separate linear stacks of
TTF molecules and TCNQ molecules. For a substantial range of temperatures,
the molecular stacks are weakly coupled and the material behaves as an array
of one-dimensional electron gases, for which the scattering rate of an electron
from an impurity is proportional to $T^{-\alpha}$, where $\alpha$ is positive if charge-density
wave (CDW) correlations are enhanced. In fact TTF-TCNQ does undergo
a CDW transition at low temperatures, so the resistivity should decrease as
the temperature is increased. However, at constant inter-molecular spacing
along a stack, TTF-TCNQ has a resistivity that increases linearly with $T$ in
the one-dimensional region where it also violates the Ioffe-Regel condition.
Thus there is not even a qualitative understanding of charge transport in this
material.

These examples illustrate the point that the transport properties of syn-
thetic metals pose a problem of fundamental importance for condensed matter
physics, inasmuch as the conventional transport theory is inapplicable. Other
examples will be given in the next section. One possible solution is that trans-
port is controlled by a quantum critical point, i.e. a critical point at zero
temperature. This could be a zero-dimensional quantum critical point, as in
the physics of the two-channel Kondo problem or a two or three dimen-
sional quantum critical point, generated by long-distance physics. More
work is needed to establish whether this idea provides a general solution of
the problem, or if other new paradigms will have to be developed. However,
at a quantum critical point, the electron-phonon coupling could be irrelevant,
so there is the possibility of explaining the lack of a phonon contribution to
$\rho$. A zero-dimensional quantum critical model of charge transport via assisted
tunnelling will be described in Sec. III.

In principle, angular resolved photoemission spectroscopy is capable of
verifying or disproving the quasiparticle picture, provided the resolution is
sufficiently good. So far the best such experiments have been carried out on
the high temperature superconductor \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta} \) and, typically, they do not validate the existence of quasiparticles in the normal state. Now, a new generation of spectrometers with much improved resolution is at hand and, in the future, we may have better input from this spectroscopy.

2 Conventional Transport Theory

2.1 Drude-Lorentz Theory

A simplified view of the central idea of the Drude-Lorentz theory of transport in metals is as follows. Consider a set of electrons of density \( n \), charge \( e \) and mass \( m \) moving with velocity \( v \). The current density is \( j = nev \). Now, if an electron experiences a constant electric field \( E \) during a mean free time \( \tau \), the velocity is \( eE\tau/m \), and the conductivity \( \sigma_0 \), defined from \( j = \sigma_0E \), is given by

\[
\sigma_0 = \frac{ne^2}{m}\tau
\]

where \( ne^2/m \) is the “Drude weight”. This simple derivation emphasises the central assumption of an electron that is freely-moving during a mean free time \( \tau \). Of course, in practice, all of the work goes into calculating the mean free time from electron-electron collisions and from the scattering of electrons by phonons and impurities. After the invention of quantum mechanics, the practical implementation of these calculations changed drastically.

An estimate of the Ioffe-Regel value of \( \rho \) gives about 0.2 to 0.3 m\( \Omega \)cm. Here, it was assumed that, in three dimensions, \( k_F^{-1} \) is about 1Å. In two dimensions, the value of \( k_F \) does not enter into the estimate in because \( n \sim k_F^2 \) and

\[
\hbar k_F^2 \tau/m = k_F v_F \tau = k_F l
\]

Thus the magnitude of \( k_F l \) is independent of issues, such as the existence of a large or small Fermi surface. In two dimensions, the length scale that enters into \( \rho \) is the spacing between planes, for which we have used 6Å, a common value in the high temperature superconductors, which are prime examples of bad metals. In general there also should be a geometric factor that expresses the shape of the “Fermi surface.”

This estimate of the Ioffe-Regel condition is widely violated. In optimally doped \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \), the resistivity in the a-b plane increases linearly with temperature from \( T_c \) up to 900\(^\circ\)K, where its magnitude is 0.7 m\( \Omega \)cm. Other high temperature superconductors have similarly high resistivities at elevated temperatures. In \( \text{Rb}_3\text{C}_6\text{O}_{9} \), the resistivity has been measured up to 520K, where the implied mean free path is 0.6Å, corresponding to a value of
\( k_F l / 2\pi = 0.03 \) In TTF-TCNQ, the room-temperature resistivity in the highly conducting direction is about 2mΩ-cm.

### 2.2 Resistivity Saturation

In normal metals with strong electron-phonon coupling, the resistivity is well described by Boltzmann transport theory at low temperatures. At high temperatures the resistivity does not violate the Ioffe-Regel condition, but saturates before doing so. For example, the resistivity of Nb₃Sn saturates at a value of about 0.1mΩ−cm.

### 2.3 Fermi Liquid Theory

In the mid fifties, Landau introduced the concept of a Fermi liquid, in which the behavior of the system is described in terms of elementary excitations out of the ground state. The excitations were assumed to be fermionic quasiparticles and bosonic collective excitations, and the idea was that, even in a strongly-correlated system, there are few such excitations at low temperatures, so transport coefficients can be evaluated by a quantum Boltzmann transport theory which includes two-particle collision processes. In this way, it is straightforward to calculate the general temperature and frequency dependence of transport and equilibrium properties, but the system-dependent magnitudes of these quantities are specified in terms of a set of phenomenological Fermi liquid parameters. This theory was devised for liquid ³He, but it soon was applied to metals and to atomic nuclei.

A major consequence of the theory is that scattering rates \( \tau^{-1} \) are proportional to \( T^2 \) at low temperatures. However, liquid ³He is a nearly-ferromagnetic Fermi liquid, and the scattering of quasiparticles from spin fluctuations gives a significant \( T^3 \) correction to the scattering rate:

\[
\frac{1}{\tau} = aT^2 - bT^3
\]

where \( b/a \) is large enough to make the \( T^3 \) term a 25\% correction at a temperature of 40mK. The scattering rate is never simply proportional to \( T^2 \), and indeed, for some time, this behavior confounded attempts to verify the predictions of Fermi liquid theory in liquid ³He. Another consequence of the nearly ferromagnetic nature of liquid ³He is that it is a fermion superfluid with pairing in odd angular momentum states.

It is particularly interesting to consider Sr₂RuO₄ in the same context. This material has a \( T \)-linear resistivity up to 110K but clean samples cross over to the Fermi liquid \( T^2 \) behavior below 30K. Sr₂RuO₄ is a superconductor.
with a transition temperature of about 1K. It has been noted that this material should have a strong Hund’s rule coupling, and that a related material SrRuO$_3$ has a ferromagnetic ground state. This leads to the suggestion that Sr$_2$RuO$_4$ may be a nearly-ferromagnetic Fermi liquid at low temperatures and to the (apparently successful) prediction that Sr$_2$RuO$_4$, like $^3$He, should have pairing in odd angular momentum states. However, it is not so clear that Sr$_2$RuO$_4$ is really nearly ferromagnetic: its resistivity is proportional to $T^2$ up to 30K, and shows no sign of the $T^3$ contribution that would characterize the scattering of electrons from ferromagnetic spin fluctuations. It may well be that new physics is involved, and that the odd-angular momentum pairing in Sr$_2$RuO$_4$ evolves from the crossover from a bad metal to a Fermi liquid as the temperature is reduced.

### 3 Charge Transport in Bad metals

#### 3.1 Common Features of Bad Metals

There are a number of common features of bad metals, although they are not universal. They are often strongly-correlated electron systems that are close to a Mott metal-insulator transition and exhibit poor screening. Frequently they are charge-transfer materials with a low carrier concentration and show charge-density or spin-density wave order or fluctuations. They often exhibit charge inhomogeneity, either self-organized (as in the high temperature superconductors) or in virtue of their structure (as in alkali-doped C$_{60}$ or organic conductors). Their resistivity is often linear in temperature $T$ or frequency $\omega$ (whichever is greater). The property that there is no energy scale for $T$ and $\omega$ is characteristic of a quantum critical point.

#### 3.2 A Model for Quasi-One Dimensional Materials

In the absence of fermionic quasiparticles, the theoretical challenge is to understand the nature of the processes which contribute to the low frequency response. This section describes one possibility – assisted tunnelling.

If there is no hopping transverse to the organic stacks in a quasi-one dimensional material, such as TTF-TCNQ, there is a flat Fermi surface. Introduction of a small transverse hopping amplitude $t_\perp$ will cause a modulation of the Fermi surface. However if $t_\perp < \pi T$, this modulation is smaller than the thermal broadening of the Fermi surface and there is no coherent motion of the charges from one chain to another. This is the definition of the one-dimensional region. Interactions actually renormalize the value of $t_\perp$ downwards and expand the one-dimensional region of temperatures. However, despite the ab-
sence of coherent tunnelling of electrons in plane wave states, there remain local hopping processes between neighboring stacks, and they enable an electron to circumvent an impurity, which would otherwise impede its motion along a given organic stack. These processes take advantage of the structural inhomogeneity and allow an electron to tunnel around the impurity. They dominate the conductivity because otherwise the central stack would become insulating at low temperature. However this is not ordinary tunnelling; as we shall see it is coupled to the low-energy electronic degrees of freedom on the adjacent stacks.

Suppose the stacks are horizontal, and that there is an impurity at the origin on a given stack. Now introduce a pseudospin variable \( \vec{\tau} \) for that stack such that \( \tau_z = \pm 1 \) designates the presence of an electron tunnelling to the right (\( \tau_z = +1 \)) or left (\( \tau_z = -1 \)) across the impurity. For an adjacent stack, there are electron variables \( \psi_{\sigma,R}(x) \) and \( \psi_{\sigma,L}(x) \) that annihilate respectively right-going and left-going electrons with spin \( \sigma \) at position \( x \). The Hamiltonian is

\[
H = H_0 + H_\parallel + H_\perp
\]

where

\[
H_0 = ivF \sum_\sigma \int dx [\psi_{\sigma,R}^\dagger \psi_{\sigma,R} - \psi_{\sigma,L}^\dagger \psi_{\sigma,L}],
\]

\[
H_\parallel = J_\parallel \sum_\sigma [\psi_{\sigma,R}(0) \psi_{\sigma,R}(0) - \psi_{\sigma,L}(0) \psi_{\sigma,L}(0)],
\]

and

\[
H_\perp = J_\perp \sum_\sigma [\tau^z \psi_{\sigma,R}^\dagger(0) \psi_{\sigma,L}(0) + H.c.].
\]

Here, \( H_0 \) is the kinetic energy, \( H_\parallel \) is a current-current coupling, and \( H_\perp \) is a backscattering interaction. It can be shown that the same model is relevant for assisted tunnelling involving several adjacent stacks.

This problem may be solved in the same way as the two-channel Kondo problem. Here the pseudospin \( \vec{\tau} \) plays the role of the Kondo spin and \( \sigma \) plays the role of a flavor label. It has been shown that the frequency-dependent conductivity at this point is given by

\[
\sigma(\omega) = \text{const.} \tan \left( \frac{\pi}{\omega} \right) \frac{\Gamma}{\omega^2 + \Gamma^2}
\]

From this equation it can be seen that

\[
\sigma(\omega) \sim \frac{1}{\omega}, \quad \text{for} \ T < \omega \ll \Gamma
\]

\[
\sim \frac{1}{\Gamma}, \quad \text{for} \ \omega < T \ll \Gamma
\]
Consequently, apart from the (large) cutoff $\Gamma$, $T$ sets the scale for $\omega$ and $\omega$ sets the scale for $T$. This behavior is a consequence of the fact that the two-channel Kondo problem is governed by a $(0+1)$-dimensional quantum critical point. (i.e. a quantum critical point in zero space dimensions and one time dimension.) Clearly, this expression gives the linear temperature dependence of the d.c. resistivity and it also implies a frequency-dependent conductivity that is proportional to $\omega$, provided $\omega > T$. This behavior will be obtained whenever there is local tunnelling assisted by low-energy degrees of freedom. In the case of alkali-doped $C_{60}$, it is unlikely that there are sufficient low-energy electronic degrees of freedom to give this behavior, but it could come from coupling to phonon modes. Other quantum critical Hamiltonians (with higher space dimension) have been proposed to account for charge transport in the high temperature superconductors, but a zero-space dimensional model has the advantage that it is more robust in disorderly materials.

A major implication of this model is that there is enhancement of composite pairing of electrons (which is equivalent to odd-frequency pairing) on the adjacent stacks in the neighborhood of the impurity. Thus, the $T$-linear resistivity is rather like a paraconductivity produced by superconducting fluctuations, although, so far, an odd-frequency pairing state has not been realised in practice.

So far, electron-electron interactions have not been included in the model. It is known that they produce logarithmic corrections to the correlation functions of the spin degrees of freedom that are responsible for the frequency and temperature dependence of the conductivity. Interestingly, it has been found that, at low temperatures, the $T$-linear resistivity either crosses over to a $\ln T$ behavior or has a $\ln T$ contribution added to it. Whether this behavior can be accounted for by adding interactions to the model is currently under investigation.

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