Invited Paper

The unconventional electrodynamics of high T_c and organic superconductors.

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ABSTRACT

The combination of lowered dimensionality and electron-electron correlations are responsible for the unusual temperature and frequency dependence of the electrical conductivity of the new superconductors. We first review the electrodynamics of two systems, $U_2Ru_2Si_2$ and Sr_2RuO_4 where conventional Fermi liquid ideas seem to work. Here transport is by free carriers with strongly renormalized masses. On the other hand the electrodynamics of the high T_c cuprates and the organic charge transfer salts is unconventional. The high T_c's show a Drude peak with an anomalous temperature and frequency dependent scattering rate for the in-plane conductivity, while normal to the planes they are almost insulating. In the organics, the transport currents are carried by a narrow collective mode coupled to phonons.

Keywords: conductivity, Drude model, superconductors, organic conductors.

1 Introduction.

In recent years the attention of the condensed matter community has turned increasingly towards materials with unconventional transport properties caused by a combination of electron correlations and lowered dimensionality. A definition of low dimensionality, for the purposes of transport properties, is a situation where there is a *qualitative* difference in the transport properties in one direction as compared to another. Thus materials where there is a large anisotropy in the band structure but where the transport can still be described by an anisotropic Fermi liquid with an anisotropic effective mass tensor, such as the conduction bands of indirect semiconductors, would not be a low dimensional material. On the other hand there are materials, high temperature superconductors, for example, where the electrodynamic properties along the copper oxygen planes are those of a good, albeit strange, metal, while the properties normal to the plane are non metallic.[1] Less studied but similar dramatic changes in the properties with current direction are seen in the organic charge transfer salts.[2]

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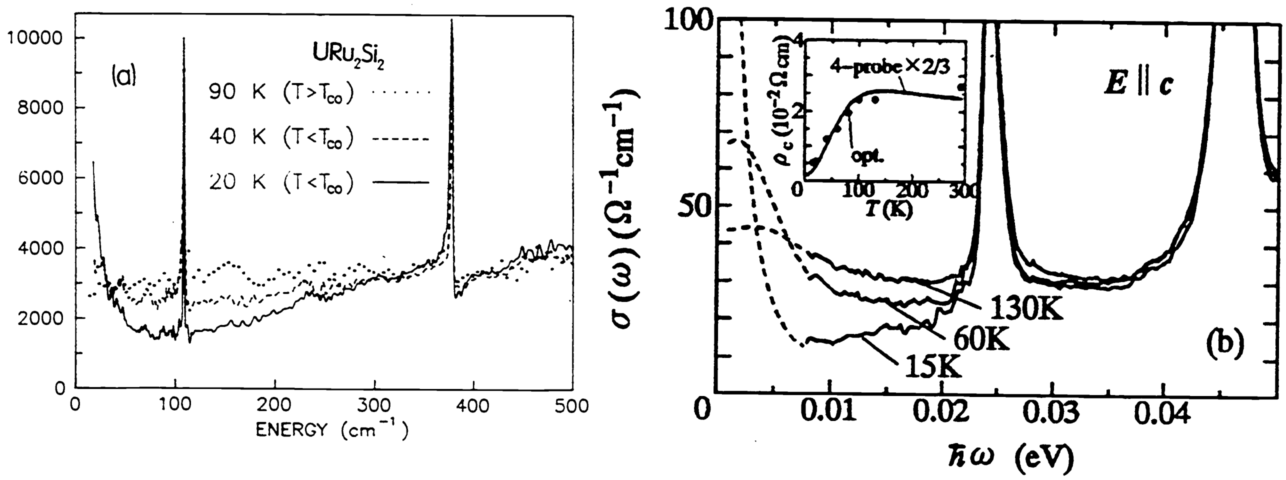

Figure 1: a) Optical conductivity in $(\Omega\text{cm})^{-1}$ of $U_2Ru_2Si_2$, b) the interplane conductivity of Sr_2RuO_4 , two systems where a narrow Drude like peak dominates dc transport at low temperatures. At high temperatures the conductivity is frequency independent.

In what follows we will review briefly the experimental data on the conductivity in several of these systems starting with two materials where there is good evidence of transport by charge carriers that interact by exchanging a low frequency excitation and where Fermi liquid ideas seem to work. One, $U_2Ru_2Si_2$, is isotropic, with a high effective mass due to magnetic interactions and the other, Sr_2RuO_4 is material that is structurally related to the anisotropic cuprates but appears to behave, with respect to c-axis transport, like a simple Fermi liquid. We then move to the high temperature superconductors where the high in-plane conductivity, to a first approximation, appears to be conventional with a Drude band with a modest mass, but we will see that there is plenty of experimental data to show that the simple theory must be questioned. The conductivity normal to the planes is highly anomalous and cannot be understood in terms of conventional transport between coherent bands. Finally we turn to the organic quasi-one-dimensional materials which display a complex set of phenomena, some of which appear to be contradictory when interpreted in terms of conventional pictures. We will see first that the on-chain transport is very anomalous and not well understood. We know even less about interchain transport, since dc conductivity is difficult to measure in this direction and little optical data exist.

A Fermi liquid, for the present purposes, is system of electrons where the transport can be described in terms of quasiparticles that are well defined and move under the influence of an electric field as free electrons but with a renormalized effective mass. The frequency dependent conductivity of such a system can be described by a generalized Drude formula with a frequency dependent scattering rate:[3]

$$\tilde{\epsilon}(\omega) = \epsilon_\infty - \frac{\omega_p^2}{\omega[m^*(\omega)/m_b][\omega + i\gamma^*(\omega)]} \tag{1}$$

where $\gamma^*(\omega) = \hbar\omega\epsilon_2/(\epsilon_\infty - \epsilon_1)$ is the renormalized damping, and $m^*(\omega)/m_b$ is the effective mass enhancement over the band mass m_b and $\epsilon(\omega) = \epsilon_1(\omega) + i\epsilon_2(\omega)$ is the dielectric function, determined from Kramers Kronig analysis of the reflectance. The quantity $\omega_p^2 = 4\pi ne^2/m_b$ is the unrenormalized plasma frequency, where n is the electron density and e the electronic charge. The area under the conductivity curve obeys the sum rule: $\omega_p^2/8 = \int_0^\infty \sigma(\omega)d\omega$. A rough rule is that if the excitations causing the scattering are well separated from the Drude peak, whose width is approximately γ^*, then the area under the Drude peak is give by the sum rule formula with ω_p^2 replaced by the renormalized value $\omega_p^{*2} = \omega_p^2 m_b/m^*$. Thus of the overall spectral weight, a fraction m_b/m^* is under the Drude peak and $1 - m_b/m^*$ under the incoherent side band corresponding to the creation of the excitations, the so-called Holstein side band, well known for the electron phonon interaction in conventional metals.[4] In applying the frequency dependent scattering rate analysis one should be careful not to include interband transitions in the dielectric function.

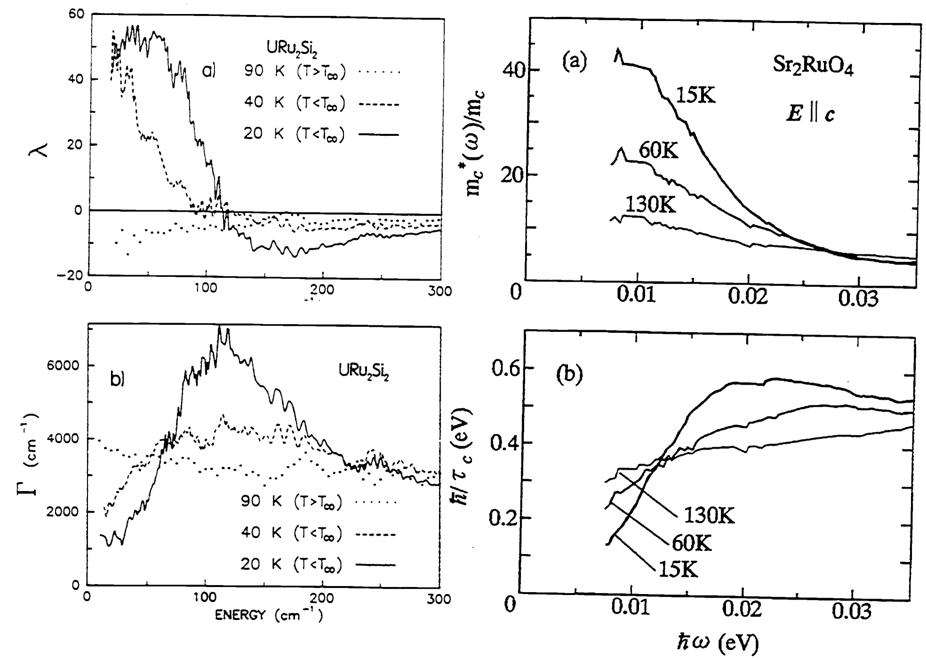

Figure 2: Left panels, a) frequency dependent effective mass and b) scattering rate of $U_2Ru_2Si_2$ and, right panels Sr_2RuO_4 . $U_2Ru_2Si_2$ is a heavy Fermion system where magnetic excitations with a characteristic energy of 100 cm^{-1} interact with the electrons. Sr_2RuO_4 is structurally related to the high T_c superconductors with interplane transport that is free-carrier like, but strongly interacting with a low lying excitation at ≈ 160 cm^{-1}.

2 $U_2Ru_2Si_2$ and $LaRuO_4$, two Fermi liquids.

A good example of a Fermi liquid system where a frequency dependent scattering rate has been evaluated is the heavy fermion compound $U_2Ru_2Si_2$.[5] The dc resistivity is nonmetallic in that it *increases* as the temperature is lowered reaching a maximum at 70 K. Below this so-called coherence temperature the resistivity drops rapidly, approximately as T^2. At high temperature, the frequency dependent conductivity shows a broad band centered of zero frequency but below the coherence temperature a narrow Drude peak appears as shown in Fig. 1a. Frequency dependent scattering rate analysis shows that the scattering rate is not constant as expected for a simple Drude model, where scattering is from static impurities, but develops a peak at approximately 100 cm^{-1} and drops dramatically below this frequency as shown in Fig 2. The area under the scattering rate curve is approximately conserved, the scattering lost at low frequencies has been shifted to the peak at 100 cm^{-1}. Inelastic neutron scattering experiments show that there is peak in the density of states of magnetic excitations that coincides with the peak in the scattering rate curve.[6]

Fig. 2a shows the effect of the strong scattering on the effective mass mass m^*. The figures for the mass enhancement are in reasonable agreement with the anomalously large specific heat that develops at low temperature in this material. Another characteristic of a Fermi liquid with a frequency dependent scattering rate is the similarity of the scattering rate and the dc resistivity, both curves rise sharply from low values to peak at a maximum frequency ω_{max} and temperature T_{max} in such a way that $kT_{max} = 0.5\hbar\omega_{max}$.

An example where the frequency dependent scattering rate formalism has been applied successfully to an anisotropic system is the recent work of Katsufuji *et al.* on Sr_2RuO_4 .[7] This material is isostructural with the high temperature superconductor $La_{2-x}Sr_xCuO_4$ with similar highly anisotropic transport properties. It is a

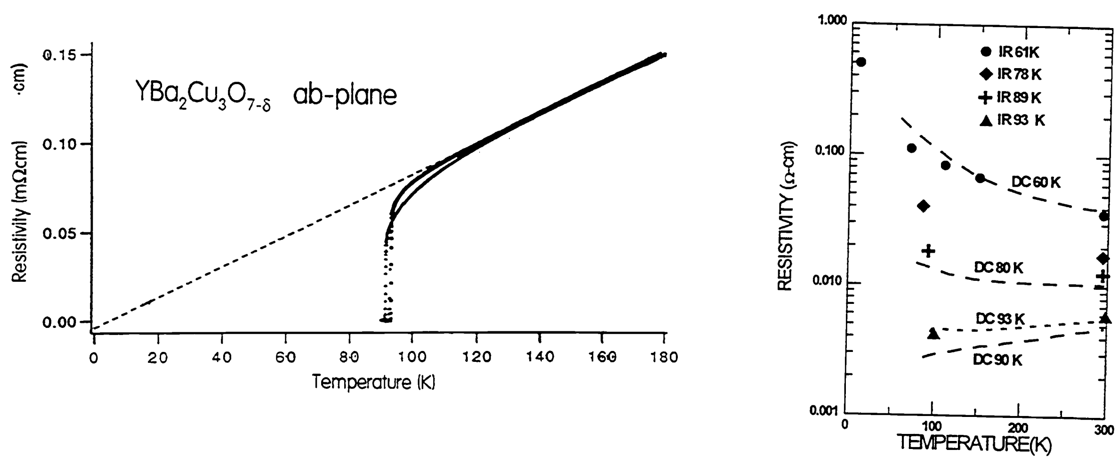

Figure 3: Left, resistivity in the ab plane single crystal of $YBa_2Cu_3O_{7-\delta}$. Above the superconducting transition the resistivity varies linearly with temperature, in the range 120 to 180 K, a dependence that extrapolates to zero resistance at zero temperature. b) c- direction interplane resistivity of a series of crystals with different oxygen dopings measured optically, symbols, and with dc techniques, solid and dashed lines. Near optimal doping the resistivity is linear but with a large constant term while the slope becomes negative in the reduced T_c materials. There is in general good agreement between the optical and dc measurements, particularly, where materials from the same source are used.

superconductor with a T_c below 1 K.[8] The c-axis conductivity at room temperature, is very low and frequency independent. In accord with this, the dc resistivity, Fig. 1b, inset, is nearly temperature independent, rising slightly as the temperature is lowered. However below 100 K the resistivity drops dramatically to a very low value of a good metal.

The optical conductivity, too, becomes metallic below 100 K. Fig 1b shows that a narrow Drude peak appears at low frequency. As the temperature is lowered below 15 K the peak moves below the low frequency limit of the experimental apparatus, but as the analysis of Katsufuji *et al.* shows, the assumption of the presence of a Drude peak at low frequency, is consistent with the dc conductivity. The frequency dependent scattering rate analysis shows that the scattering rate develops a gap-like depression below 160 cm^{-1} (0.020 eV), and in parallel to the heavy Fermion case, the area under the scattering rate curve is conserved as the temperature is lowered. As in the case of $U_2Ru_2Si_2$ the peak in the resistivity is at half of the energy of the peak in the scattering rate $kT_{max} = 0.5\hbar\omega_{max}$.

In summary, $U_2Ru_2Si_2$and Sr_2RuO_4 display many similarities in their frequency dependent conductivities but they are quite different systems. $U_2Ru_2Si_2$ is a three dimensional, nearly isotropic system where the strong frequency dependence of the scattering rate arises from strong magnetic excitations with an energy of $\approx$ 100 cm^{-1}. Above 60 K the system appears to have a diffusive conductivity, the Drude peak is very broad, and the mean free path is much less than a lattice spacing. At low temperature the material becomes a good metal with a very *narrow* Drude peak which gets narrower as the temperature is lowered. The Sr_2RuO_4 system on the other hand is an anisotropic oxide and the nature of the interactions that produce the scattering rate peak at 160 cm^{-1} is not clear at this time. While the similarity in the electrodynamics of the two systems is striking it is premature to conclude that we are looking at some generic Fermi liquid behaviour in the face of completely different scattering physics. It may well turn out that similar magnetic fluctuations are responsible for the transport scattering in both systems.

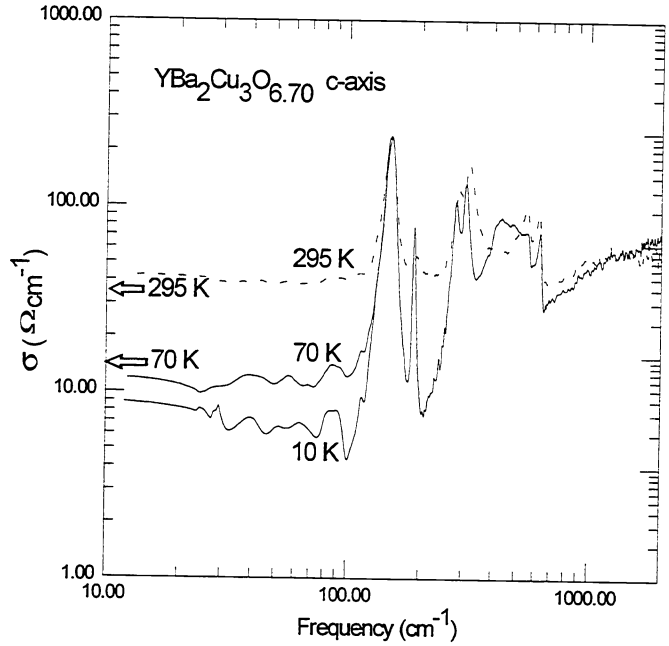

Figure 4: Interplane optical conductivity of $YBa_2Cu_3O_{7-\delta}$ at three temperatures. The superconducting transition occurs just below 70 K. At room temperature (dashed curve) the conductivity is nearly *frequency* independent apart from the peaks due to optical phonons. The high frequency conductivity is *temperature* independent but at low frequency a gap develops below 300 cm^{-1}. The temperature dependence of the dc resistivity, shown as arrows, is dominated by this gap. There is no Drude peak in this system.

3 Anisotropic transport in HTSC.

One often sees the statement that the transport of the high temperature superconductors is metallic in the direction of the planes and semiconducting in the c direction, normal to the planes. In fact the transport properties in both directions are anomalous, differing in temperature and frequency dependence from simple metals and doped semiconductors. We will look at these properties in turn.

The temperature dependence of the resistivity of a high quality $YBa_2Cu_3O_{7-\delta}$ single crystal, measured along the ab-plane, is shown in Fig 3a. The linear temperature dependence at temperatures above the superconducting transition at 92.5 K is striking. An important point to note is that the curve extrapolates close to zero resistivity at zero temperature. The dashed line shows the extrapolated line and the intercept on the temperature axis is 5 K. This, near zero intercept, is a universal property of fully doped high temperature superconductors. Below optimal doping, in the so-called spin gap region, the curves deviate from this simple form and are slightly curved.[9–11]

Attempts to explain this resistivity curve with models where the carriers form a Fermi liquid interacting with phonons or spin fluctuations fail to reproduce the zero intercept.[12,13] Instead an intercept on the temperature axis occurs at a temperature that is some fraction of the characteristic temperature of the excitation involved, the Debye temperature in the case of phonons, for example. Furthermore, the linear resistivity continues to very high temperatures and does not saturate as expected when the mean free path becomes of the order of the lattice spacing, an effect that is observed for electron phonon scattering.[14] Most importantly, in systems with a low superconducting transition temperature such as $Bi_{2+x}Sr_{2-y}CuO_{6\pm\delta}$[15] the linear resistivity variation persists to very low temperature where any phonon or spin fluctuation contributions have frozen out.

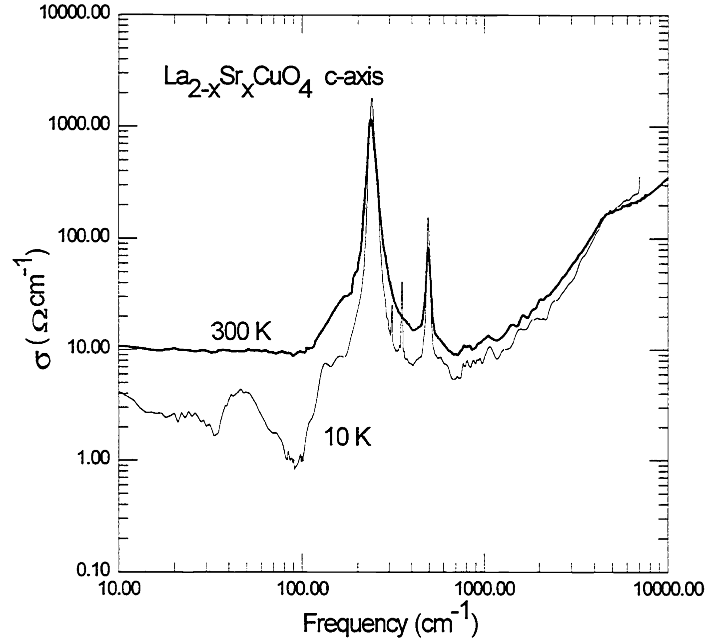

Figure 5: The c-axis conductivity of $La_{2-x}Sr_xCuO_4$. The optical conductivity is non-Drude like, being frequency independent up to 1000 cm^{-1}. There is no clear gap-like depression in the low frequency region but there is a loss of spectral weight over a very wide range of frequencies as the temperature is lowered.

The optical conductivity of the ab plane is characterized by a Drude-like peak with a width that varies linearly with temperature[16] but is accompanied by a high frequency band, the midinfrared absorption. An analysis in terms of a frequency dependent scattering rate has been used as well[17] and it shows that like the dc resistivity it is a linear dependence but the fully doped materials only.[11]

The c-axis temperature dependence of the dc resistivity is also unconventional. Fig. 3b shows data for from the work of Iye,[18] (dashed curves) Baar,[19] (dotted curve) for $YBa_2Cu_3O_{7-\delta}$ determined by four-probe resistivity measurement for samples of various oxygen dopings. At optimal doping the resistivity has a positive (metallic) slope but as the doping and T_c are lowered the slope becomes negative or "semiconducting". In some cases optical measurements provide a more accurate determination of the dc resistivity. Those are shown, for similar dopings but not the same samples, as various symbols. It can be seen that the optical determination, which is done at finite frequency but extrapolated to zero frequency, agrees well with the dc one.

The optical conductivity of the cuprates normal to the conducting planes is very low and essentially frequency independent.[20,1] Fig. 4 shows the optical conductivity in the c-direction of $YBa_2Cu_3O_{7-\delta}$ for an underdoped sample at various temperatures.[21] There are several important, unexpected features in these curves. First of all, at room temperature there is no sign of a Drude peak expected for a metal, the conductivity is essentially frequency independent. As the temperature is lowered, a pseudogap develops in the conductivity, and it is the depressed conductivity in this gap, that is responsible for the increasing resistance of the material as the temperature is lowered. The high frequency conductivity is temperature independent. This pseudogap has also been observed with other techniques such a NMR and specific heat. It's influence on the ab-plane conductivity can also be seen in underdoped materials.[10,9,11] A very similar gap occurs in the closely related double plane material $YBa_2Cu_4O_8$ that is naturally underdoped.[22]

The c-axis optical conductivity in fully doped and overdoped materials does acquire a Drude *component* with

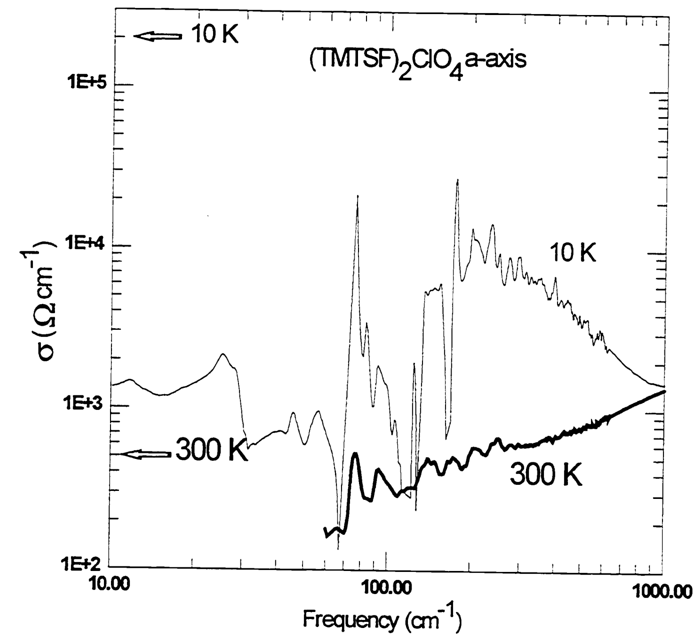

Figure 6: Optical conductivity in the conducting stack direction of the organic superconductor $(TMTSF)_2ClO_4$. The room temperature conductivity is flat and featureless, with no discernible Drude peak. The arrows show the dc resistivity for crystals from the same source. At low temperature there is a growth in the conductivity at low frequencies but dc transport and microwave measurements show that there is an extremely narrow peak in the conductivity below the frequency range shown here. At the same time several phonon lines grow in intensity suggesting that the interaction that narrows the peak is related to charge density waves.

a rather large scattering rate. This channel of conductivity is superimposed on a broad background.[23,24]

The c-axis optical conductivity of a $La_{2-x}Sr_xCuO_4$ single crystal at two different temperatures is shown in Fig 5.[25] Apart from sharp lines from optical phonons the conductivity is frequency independent. As the temperature is lowered there is an overall drop in conductivity similar to what happens in the pseudogap region of the YBCO double plane materials, but there is no sharp energy that can be identified as the gap edge where the conductivity is restored to its high temperature value. If there is a pseudogap in this material, it has an energy scale that is at least an order of magnitude higher than in YBCO.

In summary the high temperature superconductors are not ordinary anisotropic systems. In the highly conducting direction the dc resistivity is anomalous and in the c-direction, normal to the conducting planes, the conductivity is a low and frequency independent. The interplane transport is not that of an ordinary metal since no Drude peak is seen.

4 Anisotropic transport in organic superconductors.

The organic charge transfer compounds of the $(TMTSF)_2X$ family, where X is an inorganic anion, are excellent conductors at low temperature showing a metallic temperature dependence of the dc resistivity down to the lowest temperatures.[26] In contrast to this metallic behavior the low far infrared conductivity is striking. Fig. 6 shows this for $(TMTSF)_2ClO_4$where, at low temperature, there is a discrepancy of two orders of magnitude between the

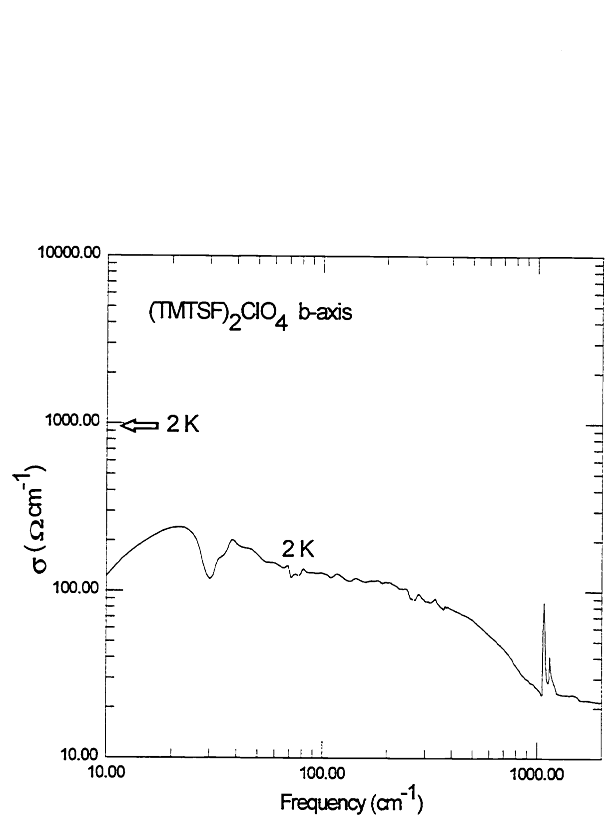

Figure 7: Optical conductivity normal to the conducting chains for $(TMTSF)_2ClO_4$. The conductivity in this direction is flat and frequency independent at all temperatures and there is some evidence of a narrow mode here too, an estimated dc resistivity point is shown by an arrow.

dc resistivity and the optical conductivity. The changeover from the low conductivity to the high conductivity occurs in the inaccessible region between microwaves and the far infrared. All the transport measurements, dc resistivity, microwave absorption and far infrared reflectance are consistent with a picture where a very narrow mode of low spectral weight and hence very large effective mass, of the order of several hundred electron masses, carries the dc transport current.[27–33]

A large anomaly in the NMR relaxation rate for $T < 25$ K[34] and the thermal conductivity suppression below 60 K[35,36] also suggest that a gap forms in the density of states due to collective effects. However there is also evidence for more conventional transport particularly in high magnetic fields. For example the observation of oscillatory phenomena in high magnetic fields[37] have generally been interpreted in terms of single particles moving in orbits.[38]

As Fig. 6. shows there is no sign of the expected Drude absorption, instead the optical conductivity is dominated by a very broad band, with $\sigma(\omega)$ *increasing* with frequency. Below 100 K, the spectral weight shifts to lower frequencies and a Drude like peak builds up with a width of the order of a few 100 cm^{-1}. However this far infrared feature does not smoothly blend into the high dc conductivity. Instead a gap develops at 170 cm^{-1} along with several phonons that grow as the temperature is lowered. These phenomena are signatures of charge density waves (CDW).

A detailed analysis of the temperature dependence of the real and imaginary parts of the optical conductivity[33] shows that the spectral weight of the narrow mode that is responsible for dc transport grows as the temperature is lowered. In this respect the behavior of the organic conductor parallels that of the two Fermi liquid materials discussed above where a growing narrow mode is responsible for the changeover from high temperature diffusive behavior to low temperature metallic behavior. However in the charge transfer compound the effect is dramatically larger: the effective mass is an order of magnitude larger and the scattering rate at least two orders of magnitude smaller.

There has been less work on the conductivity normal to the chain axis in the organic conductors but it appears that in the least conducting c-direction the conductivity has a very low value of several $(\Omega\text{cm})^{-1}$ and still exhibits a positive metallic temperature dependence. In the absence of dc resistivity measurements it is difficult to make the necessary low frequency Kramers Kronig estimates but if one takes Cooper's estimate[39] that the b direction resistivity is 50 times higher than the a direction one, one arrives at the conclusion that in the b direction too there is a low frequency mode.

Fig 7. shows the low frequency optical conductivity of $(TMTSF)_2ClO_4$ normal to the conducting stacks, in the b direction, based on the work on Ng *et al.*[40] Again, as in the a direction, the conductivity is low and essentially frequency independent. If one wants to fit this curve with a Drude model, a scattering rate of the order of a few 100 cm^{-1} would fit the data, similar to what is seen in the a direction. At low temperature a pseudogap of the order of 10 to 20 cm^{-1} seems to develop as the conductivity drops at low frequency. There appear to be no transport measurements in this crystallographic direction but from the fact that in the c-direction the material is still metallic one may assume that the b-direction is conducting as well. If that is the case we conclude that here too there is low-lying sharp mode that carries the transport current.

In summarizing the available optical data on the organic material $(TMTSF)_2ClO_4$ it appears that the charge dynamics in the ab plane is dominated by very narrow modes that have charge density wave character but there is no sign of Drude peaks associated with a large spectral weight, signatures of single particle transport in metallic systems

5 Discussion.

There is a lack of fundamental understanding of the transport properties of strongly correlated low-dimensional systems. The traditional approach is to say that the weak coupling between the conducting layers or stacks can be understood in the context of band structure where there is a low charge transfer matrix element between the planes $t_\perp$ and then proceed to deal with the problem as a three dimensional one with highly anisotropic bands. The experimental evidence presented shows that this approach fails for the c-axis transport of high temperature superconductors and the transport in all direction of the organics. In all cases the transport, in single crystals of high purity, is diffusive.

Standard quantum measurement theory has been used to account for the diffusive transport in the high T_c materials.[41] Similar ideas were advanced earlier for the organic materials.[42] The physical idea is that because of weak coupling, coherent tunnelling between the planes is a slow process, being continually interrupted by rapid in-plane scattering events, which render the interplane current incoherent. The interplane resistivity is then given by $\rho(T) \approx (t_c/t_{ab})^2\rho_{ab}$ where t_c and t_{ab} are the out-of-plane and in-plane transfer integrals respectively, and ρ_{ab} is the in-plane resistivity.

There are problems with this approach since it predicts that the temperature dependence of transport in the two directions is the same. However, in the high T_c materials the in-plane resistivity is T linear with a zero intercept whereas the c-axis resistivity is essentially temperature independent with a small linear term. To attribute the temperature independent term to impurity scattering implies a concentration of impurities such that there is an impurity in every unit cell. Microwave studies of the in-plane conductivity with doped samples show that the planar impurity level, in single crystals, is in fact very small.[43] The linear term grows relative to the temperature independent term as the doping level is increased to the so called overdoped region and may signal the onset of coherent transport in the high doping limit rather than the operation of the process of Kumar *et al.*.[23,24]

Another approach to the problem of incoherent transport is the suggestion of Anderson and his co-workers[44] that the electronic states in the planes are spin-charge separated Luttinger liquids *confined* to the planes, or the chains in case of the organics, and that single particle transport between the spin-charge separated states is essentially forbidden. The resistivity between such liquids is predicted to be almost frequency and temperature independent in the case of conduction between one dimensional chains which are expected to be, on theoretical grounds, such a Luttinger liquids.

Future experiments should include a study of the optical conductivity in a magnetic field. If the observation of oscillations in magnetoresistance with masses or the order of the free electron mass are to be interpreted in terms of onset of single particle transport above certain threshold fields then there should also be a characteristic Drude absorption with a corresponding large spectral weight.

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