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INFRARED PROPERTIES OF HIGH T_c SUPERCONDUCTORS*

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Over the past several years a coherent phenomenology of the high T_c cuprate superconductors has begun to emerge. Infrared measurements have contributed several important ingredients to this picture including: 1) the inference of a scattering rate that is linear in frequency for $\omega>T$, and of order ω, -- 2) a characteristic energy scale in the superconducting state of 500 cm^{-1} (60 meV), which can be interpreted as a superconducting pair excitation threshold or energy gap, and -- 3) evidence for very unusual temperature dependence in the vicinity of T_c. An attempt to describe these aspects of the data is presented here.

1. INTRODUCTION

Since the discovery of superconductivity in $La_{2-x}Ba_xCuO_4$ in 1986(1), a great deal of effort has been put into the study of fundamental properties of the cuprates. A primary goal of this work is, of course, to understand why certain cuprates are superconducting at very high temperatures (~100 K). Infrared spectroscopy, which covers the energy range from 10 to 10,000 K in which the characteristic energy scales relevant to superconductivity are expected to lie, can be expected to provide a very significant probe of fundamental properties. The efficacy of infrared measurements was demonstrated in the study of conventional superconductors(2-6), particularly in the early studies of the energy gap(2-5). To date, there are hundreds of papers on the infrared properties of cuprates. My goal here will not be to present a summary of this work, but instead to try to capture the essential aspects of the infrared conductivity of the cuprates, and to describe these aspects as simply as possible. In fact, I believe that the *phenomenology* of infrared properties can be described quite simply, particularly in the composition range where optimal, or nearly optimal, superconductivity occurs.

The subject of cuprates at half-filling and low doping has been discussed by Professor Uchida on Monday at this meeting, and also by Dr. Sawatzky on Friday. I will summarize this aspect only very briefly. At half-filling the cuprates are insulating due to strong electron-electron interactions. The infrared conductivity is roughly zero up to a charge transfer gap typically in the vicinity of 2 eV (16,000 cm^{-1}). With doping the strength of the charge transfer excitation diminishes, and conductivity below ~ 2 eV increases correspondingly. Strong hybridization effects appear to be relevant to understanding the rate at which the integrated infrared ($\omega\leq2$ eV) conductivity grows (Sawatzky), which is quite fast at low doping. The cuprates thus seem to be best classified as charge transfer insulators with strong hybridization.

At low doping, i.e. $x\leq0.1$ in $La_{2-x}Sr_xCuO_4$, the infrared conductivity typically has a local minimum near $\omega=0$, grows with frequency to a maximum between about 2000 and 8000 cm^{-1}, and then decreases slowly with ω up to the remnant charge transfer threshold. This behavior is clearly seen in the $La_{2-x}Sr_xCuO_4$ data of Uchida et al.(7,8), and in data from Y doped $Bi_2Sr_2CaCu_2O_{8-y}$ samples of Terasaki et al.(9,10).

This low doping regime has also been examined by Tokura et al. for the electron doped superconductors, and most recently by Thomas et al. (11). Possible reasons for the form of $\sigma(\omega)$ in this low doping regime, including aspects of the correlated band-structure, and suppression of d.c. conductivity by strong localization effects have been discussed by others at this meeting (e.g. Uchida, Sawatzky, Hanke). In this talk, I will concentrate instead on the moderate doping ($\delta\sim.15$-$.25$) regime where high Meissner fraction superconductivity occurs, and where the phenomenology of the conductivity seems to be rather simple. I will try to demonstrate this simplicity of form as well as the highly unusual nature of the infrared response.

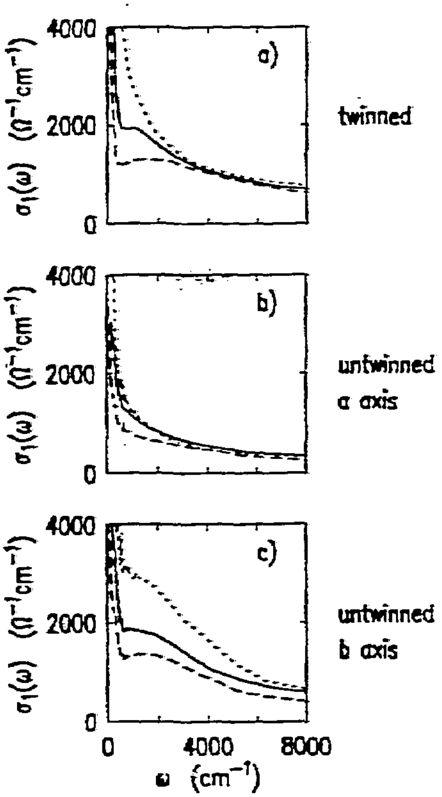

FIGURE 1
The normal state conductivity (at T~100-200 K) of twinned $YBa_2Cu_3O_{7-y}$ crystals (a), and the resolution of the conductivity into $\hat{a}$ and $\hat{b}$ components, (b) and (c), is shown. The data in (a) is from ref. 18; the data in (b) and (c) is from ref's 19 & 20. The T_c's for these samples are approximately 90 K (dotted curve), 80 K (solid curve) and 55 K (dashed curve).

2. NORMAL STATE

Much of the infrared work on cuprates has involved $YBa_2Cu_3O_{7-y}$, which is a very good material to study because of the apparently high degree of electronic homogeneity. Studies of this material can be complicated by the presence of conducting chains. Measurements of twinned crystals of $YBa_2Cu_3O_{7-y}$(12-18) yield a conductivity that involves contributions from both chains and planes. In figure 1a the normal state conductivities measured for three twinned crystals with T_c's of 90, 80 and 50 K, respectively, from Orenstein et al.(18) are shown. For the samples with $T_c\simeq80$ K and 50 K, the conductivity exhibits a local maximum in the 1000 to 4000 cm^{-1} range, in addition to the peak at $\omega=0$. In parts b and c of figure 1, we show the conductivity of several untwinned crystals of $YBa_2Cu_3O_{7-y}$ with similar T_c's (92, 80, and 56 K) from Schlesinger et al.(19) and Rotter et al.(20). In part (b) the infrared radiation is polarized along the $\hat{a}$ axis, thus one obtains the pure conductivity of the CuO_2 planes; whereas in part (c) the polarization is along the $\hat{b}$ axis (parallel to the CuO chains) and, like the twinned data, is enhanced due to the presence of the chains. One thus finds that for the pure CuO_2 planes response, the conductivity well above T_c has a very simple form in $YBa_2Cu_3O_{7-y}$, with no local maximum for $\omega<8,000$ cm^{-1}.

In principal, two structural aspects are of potential relevance to understanding the a-b anisotropy of $\sigma(\omega)$ in $YBa_2Cu_3O_{7-y}$. $YBa_2Cu_3O_{7-y}$ has chains oriented parallel to $\hat{b}$, and the in-plane Cu-O bond length is about 2% longer in the $\hat{b}$ direction than in the $\hat{a}$ direction. Rather generally, a longer Cu-O bond length would lead to a lower conductivity, thus the observed anisotropy is opposite in sign to that expected from the bond length difference. Instead, both the higher conductivity along $\hat{b}$, and the greater complexity observed in this direction, indicate that the conductivity is enhanced by the presence of chains along $\hat{b}$. Of central importance, however, is that the conductivity along the $\hat{a}$ direction is quite simple for all values of T_c considered here ($T_c\gtrsim55$ K).

One might ask whether the same simple form is followed in other high T_c superconductors. To address this question, in fig 2 the conductivity in the normal state for $La_{2-x}Sr_xCuO_4$ (x≃0.15), $Bi_2Sr_2CaCu_2O_{8-y}$ (T_c≃90), and $YBa_2Cu_3O_{7-y}$ (T_c=92 K and T_c=80 K) are shown. The $La_{2-x}Sr_xCuO_4$ data is from Uchida et al.(8), the $Bi_2Sr_2CaCu_2O_{8-y}$ data is from unpublished transmission measurements of Mandrus et al. (Stoneybrook), and is equivalent to the Terasaki data(10). The $YBa_2Cu_3O_{7-y}$ data are from Schlesinger et al.(19) and Rotter et al.(20), respectively. One sees that for all these samples the form of the conductivity is quite similar and quite simple. On a log-log plot, $\sigma_{1n}(\omega)$ can be seen to fall like $1/\omega^n$, with n of about 0.7-1. This is a much slower rate of decrease than the conventional Drude form, for which n=2 for frequencies greater than the scattering rate.

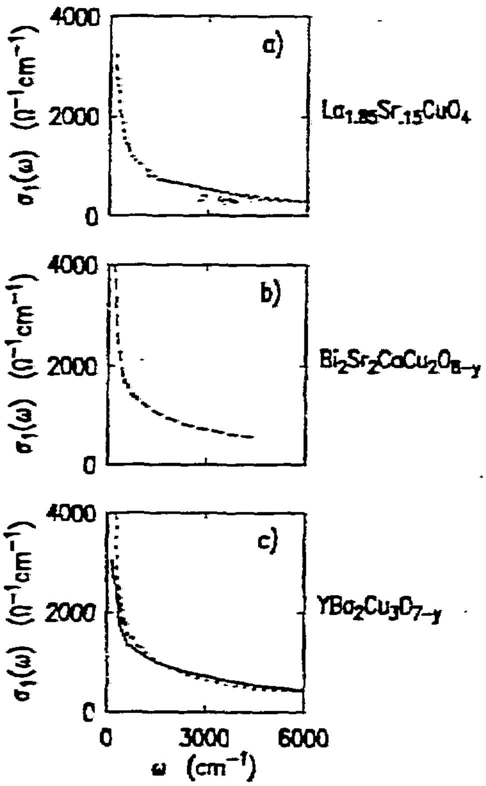

FIGURE 2
The normal state conductivities of $La_{1.85}Sr_{.15}CuO_4$ (a), $Bi_2Sr_2CaCu_2O_{8-y}$ (b), and $YBa_2Cu_3O_{7-y}$ (c) are shown.

Let us now consider interpretations of the normal state data. One approach, has been to fit the conductivity using a conventional Drude term and a sequence of Lorentz oscillators. In this approach one often associates the Drude term with the intraband contribution to $\sigma(\omega)$, while the Lorentz oscillators are associated with interband excitations. One can indeed fit the normal state conductivity for $\omega \lesssim 5000$ cm^{-1} using a Drude term and 3 additional Lorentz oscillators. Such a fit, using parameters very similar to those of Kamaras et al.(21) is shown in figure 3. Given the number of parameters available (eleven), it is not surprising that this provides a reasonably good fit to the data. The additional step of collapsing the (entire) Drude term to a Dirac δ-function at $\omega=0$ to fit the superconducting state data, which has also been considered(21), does not work well (dashed line, figure 3).

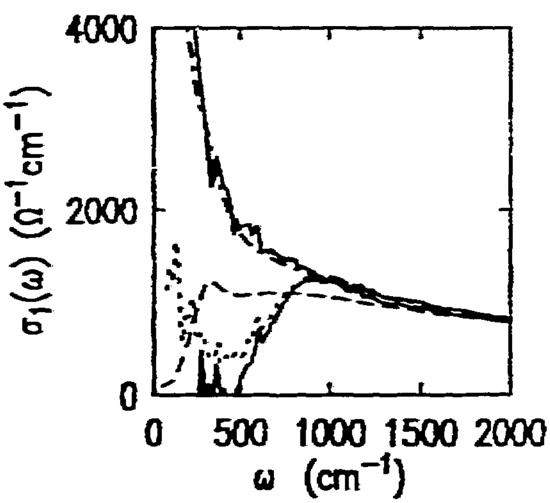

FIGURE 3
Attempts to fit the normal and superconducting states using a sequence of Lorentz oscillators and a Drude term is shown (after Kamaras et al.(21)). The dot-dashed curve is the fit to the normal state data (upper solid curve). The dashed curve is a (failed) attempt to fit the superconducting data by reducing the width of the Drude term to 0, i.e., collapsing it to a zero frequency δ-function. The data are from both reflectivity(19) (solid curves) and absorptivity(27) (dotted curve), as discussed in the following section (see also fig 7).

Another approach is to treat all the conductivity below, for example, $\sim 2000\ cm^{-1}$ as a single entity, and to then represent the electronic response in terms of a frequency dependent mass and scattering rate, as shown in fig. 4. One then finds that the scattering rate is proportional to the frequency for $\omega \gtrsim T$ (ref. 19). This approach reveals a fundamental relationship between the temperature dependence of the resistivity (linear in T), and the frequency dependence of the scattering rate (linear in ω). This approach also allows one to readily see a connection between the linear scattering rate as a function of frequency (energy) and a linear photoemission linewidth as a function of energy, which has also been widely discussed(22-24). One probes single particle response, while the other probes two-particle response, but both appear to be reflecting the same physics. An apparently related phenomenon is the broad electronic background observed in Raman scattering (23). Together these measurements present a coherent picture of a highly unusual (non-Fermi liquid) normal state phenomenology.

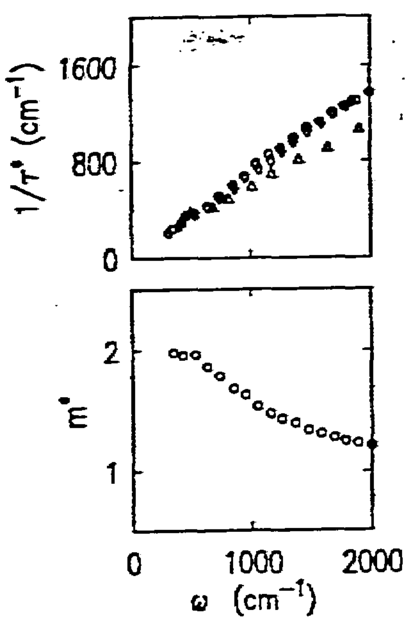

FIGURE 4
Scattering rate (renormalized) vs frequency is shown for $La_{1.85}Sr_{.15}CuO_4$ (diamonds), $Bi_2Sr_2CaCu_2O_{8-y}$ (triangles) and $YBa_2Cu_3O_7$ (cirles). Also shown in part (b) is the mass enhancement as a function of frequency for $YBa_2Cu_3O_7$.

For the $T_c \sim 90$ K superconductors, the phenomenology of the normal state can be summarized quite simply. At $\omega = 0$, we know from d.c. resistivity measurements that $\sigma_{1n}(\omega)$ is proportional to the 1/T. At higher frequency the conductivity drops with frequency like $1/\omega^n$, with $n \simeq 0.7$-1. The temperature dependence of $\sigma_{1n}(\omega)$ is primarily confined to the frequency range $\omega \lesssim 2kT$. (For T=150 K, e. g., this means $\omega \lesssim 200\ cm^{-1}$.) These essential aspects of the normal state conductivity are illustrated in fig 5. Finally, we emphasize that unlike conventional metals, where the scattering rate establishes a characteristic energy scale in the normal state, the cuprates with $T_c \sim 90$ K show no characteristic energy scale in the normal state, other than kT.

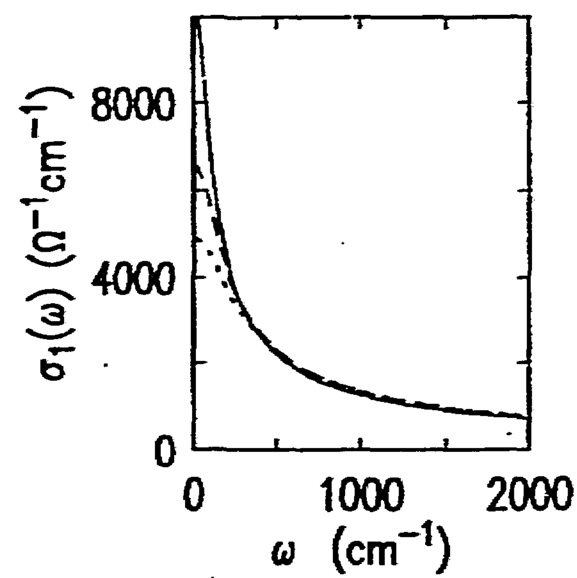

FIGURE 5
Normal state temperature and frequency dependence of the infrared conductivity (schematic) at 100 K (solid), 200 K (dashed) and 300 K (dotted). At low ω ($\lesssim 2$ kT), $\sigma_{1n}(\omega)$ increases with decreasing T. The sum rule is satisfied by a corresponding very modest decrease in $\sigma_{1n}(\omega)$ extending over a very wide frequency range (3 kT$\lesssim \omega \lesssim$100 kT).

3. SUPERCONDUCTING STATE

For the T_c~90 K superconductors, a characteristic energy scale begins to emerge near T_c, and is quite evident deep in the superconducting state. This can be seen both in ratios (fig 6) and in the absolute conductivity (fig. 7). Figure 6 compares reflectivity, transmission and conductivity ratios (superconducting state divided by normal state), for conventional and cuprate superconductors. These data are quite suggestive (of a superconducting energy gap), in that, for cuprates with T_c~90 K, the changes in the low frequency spectra associated with superconductivity are quite similar to those seen in conventional superconductors(2-5) and $Ba_{1-x}K_xBiO_3$(25).

From the ratios shown in fig 6 characteristic energies corresponding to about $2\Delta/kT_c \simeq 4$ have been inferred for the conventional superconductors, Pb, V, and Nb (2-5). From the T_c~90 K cuprates, however, the characteristic energy inferred from these ratios is about 500 cm^{-1}, which corresponds to $2\Delta/kT_c \simeq 8$. (This is where the conductivity threshold occurs.) As an interesting side-issue, we note that the displacement of the peak in T_s/T_n to higher frequency than 500 cm^{-1} (~600-700 cm^{-1}) can be shown to result from the unusually slow decrease of $\sigma_1(\omega)$ in the normal state. This effects $\sigma_2(\omega)$ (through causality) in both the normal and superconducting states and causes the peak in T_s/T_n to occur above the frequency of the conductivity threshold(26). This is different from conventional superconductors where the peak in T_s/T_n occurs very close to 2Δ (3).

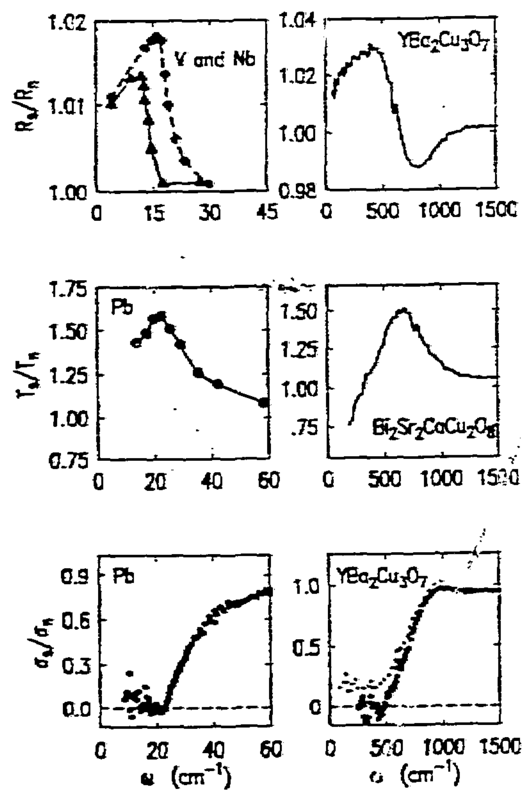

FIGURE 6
Comparison of changes due to superconducting transition in certain conventional superconductors(3-5), and cuprates with T_c~90 K. The $YBa_2Cu_3O_7$ refelctivity ratio is from references 12, 16 and 17. The $Bi_2Sr_2CaCu_2O_{8-y}$ transmission ratio is from unpublished data of Mandrus et al.. The $YBa_2Cu_3O_7$ conductivity ratios are from untwinned crystals using the reflectivity (solid) and absorptivity (dotted curve) data of Schlesinger et al.(19) and Pham et al.(27).

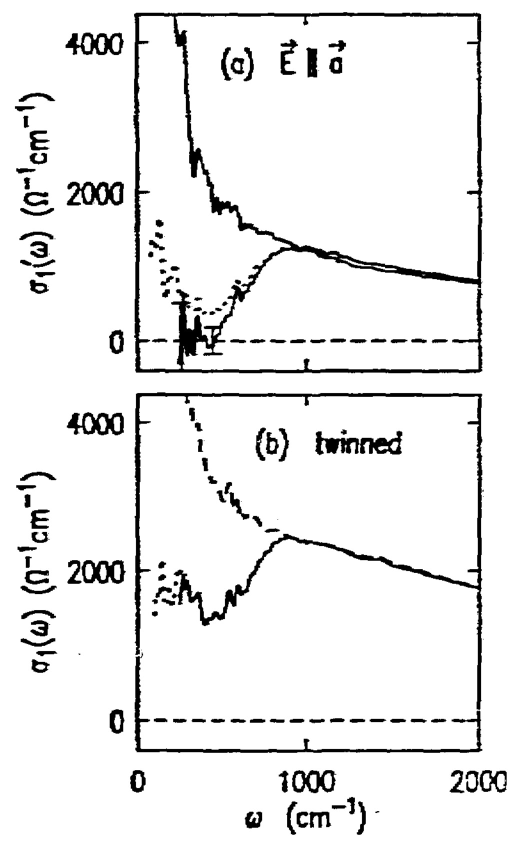

FIGURE 7
a) The conductivity of the CuO_2 planes in $YBa_2Cu_3O_7$ in the normal (upper curve, 100 K) and superconducting state (30 K) are shown from a untwinned $YBa_2Cu_3O_7$ crystal(19) (part a). The dotted curve is from absorptivity data (27). In (b) the same quantities are shown for twinned $YBa_2Cu_3O_7$ (17,28).

Figure 7 shows superconducting state conductivities obtained from reflectivity(19,17) and from absorptivity measurements(27,28). In part (a) the data are the pure conductivity of the CuO_2 planes, as measured in an untwinned crystal(19,27), while in part (b) conductivity data from a twinned crystal(17,28) are shown. One can see that significant progress has been made in this low frequency range by studying untwinned crystals, in that the conductivity is now getting much closer to zero in the superconducting state than it did for the twinned crystals. In part (a), the solid line is from reflectivity data(19) and the dotted curve is from absorption measurements(27). The data are similar in that both show clearly the conductivity threshold at $\sim 500\ cm^{-1}$, and both show no evidence for any other characteristic energy scale within the range of the data (down to $\sim 80\ cm^{-1}$ for absorptivity).

From the reflectivity measurement the level of the conductivity is consistent with zero below $500\ cm^{-1}$ in the superconducting state. From the absorption measurement a conductivity level of about 20% of $\sigma_{1n}(\omega)$ can be inferred. It is not clear at the present time if this absorption in the superconducting state is intrinsic, or if it is due to poor surface quality. The Maryland group is currently working on improved surface preparation and measurement techniques to address this important question. To date, the crystals which have been studied have had as-grown or polished-in-air surfaces, which are expected to be less than ideal. Difficulties with tunneling and photoemission measurements due to surface quality, especially with $YBa_2Cu_3O_{7-y}$, are well known. To some extent one can get away with studying less than ideal surfaces with infrared because the penetration depth is so large (~150 nm), however, with high precision measurements the issue of surface quality becomes increasingly relevant even for infrared measurements.

People sometimes ask if it is true that the cuprates are "too clean to see the gap". This idea is predicated on the expectation that the intraband contribution has a conventional Drude form (falling like $1/\omega^2$), and that there is therefore no significant intraband contribution to $\sigma_1(\omega)$ at the gap frequency(21). This point of view does not seem to have any relevance for the cuprates, simply because the normal state $\sigma_1(\omega)$ does not fall so rapidly, and thus there is a great deal of conductivity even at $500\ cm^{-1}$ as seen in fig 7. In fact there is much more conductivity at $\sim 500\ cm^{-1}$ than one would expect based on a conventional Drude picture, and the changes in the conductivity below T_c are in fact quite large, as seen in fig 6 and 7. These changes cannot be explained by collapsing a Drude term to a Dirac δ-function, as acknowledged in the Erratum for reference 21, and as shown here in fig 3.

I believe that the perception of controversy in the infrared field may be significantly greater than the reality. Often the disagreements between different groups are based in semantics rather than real differences in data or physics issues. At this meeting there seems to be widespread agreement that the way to describe the normal state data for good superconductors is in terms of a scattering rate that is linear in ω(19). This point of view was presented by Uchida on Monday for $La_{1.85}Sr_{.15}CuO_4$, and in the present talk for the $YBa_2Cu_3O_{7-y}$ and $Bi_2Sr_2CaCu_2O_{8-y}$ data. This approach has also been adopted by Batlogg (AT&T) in his summary of this conference and in a recent Physics Today article.

Regarding the energy gap controversy, I would like to quote from the recent Phys. Rev. B article(18) on infrared properties by Orenstein, Thomas, Millis, Cooper, Rapkine, Timusk, Schneemeyer, and Waszczak, which states that "*the comparison of one- and two-particle spectroscopies strongly suggests the existence of a gap of magnitude 50-60 meV in superconductors with T_c~90 K.*" This is essentially identical to our point of view regarding the gap. In our work we have found that for superconductors with T_c~90 K, there is an absorption threshold at ~60 meV ($500\ cm^{-1}$), which can be readily interpreted as an energy gap or pair excitation threshold in the superconducting state. A similar point a view is also taken by Renk et al.(29), who in very careful far-infrared and microwave studies of $YBa_2Cu_3O_{7-y}$ films, find "*...evidence for a superconducting gap at $2\Delta/kT_c \simeq 7$*

(~55 meV)..", and "..no other feature that may be due to a second energy gap at lower frequency, as suggested earlier.".

The absence of evidence for a lower gap has also been found in the absorption studies of Pham, Drew et al.(27,28) and of Miller, Richards et al.(30). For the direction perpendicular to the CuO_2 planes however, there is evidence for a lower characteristic energy scale in the superconducting state(16), which is roughly about 100 -200 cm^{-1}. Thus far the nature, existence or magnitude of a superconducting gap for the chains, remains an unresolved issue. Some Raman data from '124' (Heyen et al.(31)) suggests a very low characteristic energy (~100 cm^{-1}) for the chains.

We have discussed so far two aspects of the infrared data. One is the normal state conductivity which drops unusually slowly as a function of ω and can be represented in terms of a scattering rate linear in ω. The other is the superconducting state conductivity, which exhibits an energy scale of ~500 cm^{-1}, which is unusually large compared to T_c. (For $T_c \simeq 90$ K, $2\Delta/kT_c \simeq 8$.) For the CuO_2 plane response, this is the only characteristic energy scale in the infrared data, thus if one believes that there is an energy gap, it is almost certainly associated with the 500 cm^{-1} (60 meV) feature. Comparisons to data from conventional superconductors (fig 6) show that this feature presents itself in very much like an ordinary superconducting gap.

A further aspect of the infrared data which we have not been able to discuss here due to space limitations is the temperature dependence of this gap feature. In particular one finds that this is quite unusual in that the gap goes away near T_c by "filling-in"(19), but does not collapse to lower energy as in the conventional BCS theory. This has been discussed in references 17 & 19, and in more detail in ref 32 and 33 where a two-fluid approach has been considered. In ref 33 comparison has been made between the temperature dependence of the infrared conductivity, and that of the relevant planar NMR (or NQR) relaxation rates. These two quantities (as well the Knight shift) have remarkably similar temperature dependencies. This comparison has also been made recently for reduced T_c (~60 K) $YBa_2Cu_3O_{7-y}$ (Rotter et al.(20)), where the temperature dependence of these quantities is even more unusual. Possible implications of these data including their relationship to a normal state spin-gap scenario (discussed by Fukuyama, Lee, Rice and others, at this conference and at the Toshiba school in Kyoto), are discussed in reference 20. Overall, much of the infrared, NMR and neutron data suggest a highly unusual phenomenology above and below T_c, in which the onset of phase coherence in cuprates may lie somewhere between the gap opening transition of BCS theory, and Bose condensation of preformed pairs.

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