Analysis of the $a-b$ plane optical conductivity $\sigma_{ab}$ for both twinned and untwinned \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ as a function of temperature and doping shows that below a well defined temperature $T^*$, a dip in the spectrum systematically appears separating the infrared charge excitation spectrum into two components with distinct energy scales. The change from monotonic behaviour in $\sigma_{ab}$ is found to be concurrent with the onset of phonon anomalies in Raman and infrared spectra below $T^*$. The optical data are suggested to be evidence for the appearance of an inhomogeneous distribution of carriers rather than the opening of a simple gap for charge excitations below $T^*$, an interpretation which is consistent with recent angle-resolved photoemission and electronic Raman spectra. We find that the behaviour below $T^*$ and the absence of any anomalies at $T_c$ can be interpreted assuming a Bose-Einstein condensation of preformed pairs.

Following theoretical ideas on inhomogeneous charge carrier distributions and phase separation in cuprate superconductors \cite{1,2}, significant experimental support has recently been accumulating for a two-component charge carrier picture, particularly in the underdoped region of the phase diagram of these materials. Structural evidence from extended X-ray absorption fine structure (EXAFS) \cite{3} and neutron pair distribution function analysis (PDF) \cite{4} as well as NMR, NQR and Mossbauer \cite{5} spectroscopies \cite{6} indicate that below a certain cross-over temperature $T^*$, the Cu-O planes may contain stripes of carrier-rich and carrier-poor regions on length scales typical of the superconducting coherence length $\xi$. These features apparently have quite fast dynamics \cite{11} making the textures observable only with spectroscopies with sufficiently short characteristic timescales. Although initially the NMR data suggested that the underdoped state may be characterized by a gap primarily for spin excitations, recent angle-resolved photoemission (ARPES) \cite{12} and time-resolved optical photomodulation data \cite{13} suggest that some kind of a gap may also be present - at least for parts of the Fermi surface - for charge excitations as well. The appearance of such a pseudogap above $T_c$ appears to be quite a general phenomenon and is observed also in many other systems, including the superconducting bismuthates like Ba$_{1-x}$K$_x$BiO$_3$ \cite{4}, heavy Fermion superconductors \cite{13} and possibly even in the roton excitation spectrum of LHe \cite{10}, implying a possible common underlying reason for its existence in all these systems.

However in the case of the cuprates, the data from a key experiment - infrared optical spectroscopy - has not yet been systematically discussed in terms of the above-mentioned picture. Although the existence of a "pseudogap" in the out-of-plane optical conductivity $\sigma_\|$ has been investigated in some detail \cite{17,18,20}, the in-plane optical data has so far been discussed mainly in terms of a single-component framework of an extended Drude model with temperature- and frequency-dependent scattering rate $\tau(\omega,T)$ and carrier mass $m^*(\omega,T)$ \cite{26,27} and no systematic analysis of $\sigma(\omega,T)$ as a function of doping and temperature has yet been performed.

It is the purpose of this paper to present an analysis of the available $a-b$ plane optical conductivity data in terms of the two-component picture. We concentrate primarily on YBa$_2$Cu$_3$O$_{7-\delta}$ where the data are most complete and the contributions to the optical conductivity arising from the chains and planes can be identified from $a-b$ anisotropy measurements of $\sigma_1(\omega,T)$ \cite{28}. Our analysis of the doping and temperature dependence of the optical conductivity are performed on published data and unambiguously show that the in-plane optical conductivity qualitatively changes at a characteristic temperature $T^*$ from a single-component spectrum above, to one with (at least) two components below $T^*$. These findings together with enumeration of various phonon anomalies in the Raman and infrared spectra, collected from the available data apparently confirm the two-component paradigm of the normal state in the underdoped materials below $T^*$, and define the temperature and frequency dependence of the excitations in the two-component state.

In common with most near-optimally doped cuprate samples, the real part of the room temperature optical conductivity along the crystallographic $a$-axis $\sigma_{1a}$ of YBa$_2$Cu$_3$O$_{7-\delta}$ below $\omega \sim 400$ cm$^{-1}$ shows a monotonic decrease with increasing frequency reminiscent of an inverse law $\sigma(\omega) \sim 1/\omega$. Above this frequency the inverse law fails and an additional spectral weight is necessary to describe the spectra \cite{29}. Below room temperature (but still above $T_c$) a dip ubiquitously appears in the range 400 - 800 cm$^{-1}$ which becomes more pronounced with further cooling and
eventually below $T_c$ the residual conductivity shows only a characteristic mid-infrared (MIR) peak at around 1000 cm$^{-1}$ which again is a feature common to all the cuprates. To analyse this behaviour of $\sigma_1(\omega, T)$ more quantitatively, we have selected an arbitrary criterion to define the temperature $T^*$ at which $\sigma_1(\omega, T)$ departs from monotonic behaviour and the first derivative $\sigma'(\omega)$ shows a maximum (i.e. a point of inflexion appears) as shown schematically by the arrow in the Figure 1. Data for a number of YBa$_2$Cu$_3$O$_{7-\delta}$ samples with different $T_c$'s and $\delta$ were collected and the results of the analysis are plotted as a function of $T_c$ in Figure 2. Because there are usually not many spectra at different temperatures available, the data are necessarily plotted with a range of temperatures indicated as error-bars. Nevertheless, the correlation between the temperature $T^*$ where the spectrum departs from monotonic behaviour and $T_c$ is quite evident and the relation appears almost linear in YBa$_2$Cu$_3$O$_{7-\delta}$ over the whole range of $\delta$.

To complement the analysis on $\sigma_1$, we have systematically collected infrared and Raman phonon spectra on YBa$_2$Cu$_3$O$_{7-\delta}$ with different $\delta$ and determined the temperatures $T_{\text{anomaly}}$ at which phonon anomalies appear. To expand the data set somewhat we have also used the published data on phonon anomalies in the 124 and 247 structures, which have similar optimal $T_c$ as 123 [30]. IR phonons anomalies are observed above $T_c$ for phonons involving c-axis displacements of Y, Ba, O1 (chain), O2/O3, O4 and Cu1(chain) ions [24,32] in 123 system and of O1 (chain) and O2/O3 ions in 124 system [33,34]. Among Raman active phonons it is interesting to note that the 340-cm$^{-1}$ phonon shows little or no anomalies above $T_c$ in underdoped region of doping [33,34]. On the other hand the related in phase $A_g$ O2/O3 planar buckling phonon shows little or no anomalies above $T_c$ in underdoped region of doping [33,35]. In the 124 system [36] anomalies above $T_c$ in underdoped region are reported also for the 108-cm$^{-1}$Ba phonon, 256-cm$^{-1}$ Cu1 phonon and 501-cm$^{-1}$ O4 phonon. There is no clear data on whether such anomalies for Ba and O4 phonons exist also in YBCO-123.

The criterion for determination of the temperatures $T_{\text{anomaly}}$ was a discontinuous change in the phonon frequency and/or linewidth. A plot of $T_{\text{anomaly}}$ as a function of doping is shown in the insert to Figure 2 and as a function of $T_c$. The larger scatter in the data in this case is partly due to the ambiguity in determining the exact temperature where a phonon anomaly occurs. Despite this, it can be seen, that in virtually all cases phonon anomalies appear below $T^*$. Depending on $\delta$, $T_{\text{anomaly}}$ seems to vary such that in the ortho-II phase of YBCO (70K < $T_c$ < 93K), $T_{\text{anomaly}} \lesssim T^*$, while in the Ortho-I phase (0 < $T_c$ < 70 K), $T_{\text{anomaly}} \ll T^*$. For optimal doping there is no sign of any phonon anomalies above $T_c$, so we assume $T_{\text{anomaly}} \approx T_c$. Unfortunately, at present there is insufficient data to be able to correlate the behaviour of particular phonons with different values of $T_{\text{anomaly}}$ and there is a clear need for such a study in the future.

For completeness, the $T^*$ data from optical conductivity are plotted as a function of oxygen content in Figure 3 together with $T_c$. These data for $T^*$ obtained from $\sigma_{ab}$ are in good agreement with data on $\sigma_c$ [12,24] and with other data from a variety of experimental techniques on YBCO, including ARPES, DC transport and NMR, where it is often called a spin gap temperature $T_{sg}$. In the present case, the optical conductivity experiments measure only dipole transitions, and $T^*$ therefore signifies the pseudogap opening for charge excitations. The appearance of a pseudogap for spin excitations will thus necessarily follow at the same temperature. (The converse is not necessarily true). Although there appears to be an apparent spread of values of $T^*$ and $T_{sg}$ in the literature, suggesting the possible existence of two pseudogaps appearing at different temperatures, when it is taken into account that very different criteria are used to define $T^*$ by different authors, and that different spectroscopic techniques average over momentum in different ways, data on YBCO do not appear to support the two gap hypothesis at present. Careful examination of the optical conductivity data for $\sigma_{ab}$ and $\sigma_c$, particularly in comparison with the data on the increase in NMR $T_1$ relaxation time (usually expressed as a drop in $1/T_1T$ [22,21]), NMR Knight shift $K$ [21,22], neutron scattering [24] and specific heat data [25] does not yield any systematic difference in $T^*$. However, with significantly improved sample characterization in the underdoped state and a well-defined common criterion for defining $T^*$ the existence of two distinct, closely spaced gaps cannot be excluded.

Let us now turn to a discussion of the presented data in the general framework of the inhomogeneous phase models. Commonly, the state below $T^*$ is suggested to consist of carrier-poor and carrier-rich regions. The optical response is expected to reflect this, the existence of identifiable different spectral features below $T^*$ and a single component above $T^*$ is in clear accordance with these models. Below $T^*$, the low-frequency spectrum - which is assumed to be due to itinerant charges - is strongly temperature-dependent, has no identifiable spectral signatures and is described well phenomenologically by an extended Drude model (albeit over a small spectral region). As a corollary, the MIR would be expected to be due to the excitation of bound localized carriers to the itinerant carrier 'Fermionic' phase in addition to non-adiabatic hopping processes within the insulating phase itself as will be discussed below.

Examining the behaviour of the MIR feature as a function of $\delta$ in more detail, we have plotted the residual conductivity at the peak in the MIR (usually in the range 1000 - 1300 cm$^{-1}$) below $T_c$ as a function of doping in Figure 4. The data extracted from Rotter et al. [28] are of particular relevance because they are from untwinned single crystals (also for lower $\delta$). We find an unambiguous increase in $\sigma_1^{\text{MIR}}$ with increasing $\delta$. Comparing the $\sigma_1^{\text{MIR}}$ with
\( \sigma_{\text{MIR}}^{\text{MIR}} \) shows that the in-plane conductivity \( \sigma_{\text{ab}}^{\text{MIR}} \) shows very similar systematics as the total conductivity \( \sigma_{\text{ab}}^{\text{MIR}} \). The ratio \( \sigma_{\text{MIR}}^{\text{ab}} / \sigma_{\text{MIR}}^{\text{ab}} = \zeta \) appears to be only weakly dependent on \( \delta \), indicating that the increasing O doping gives rise to increasing conductivity in the chains and planes on an almost equal footing. Moreover, the ratio between the total and chain conductivity is \( \zeta = 1.4 \) to 1.8 and is surprisingly similar to the anisotropy of the DC conductivity \( \sigma(0) \) which suggests that O doping results in an increase in conductivity of both the Drude and MIR parts equally (see Figure 4).

The spectral shape of the MIR is similar (but not identical) to the photoinduced polaronic features observed in the insulating phase in YBCO, and is virtually indistinguishable from the MIR response of the insulator PrBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) \[13,14\] (including anisotropy). This, together with clearly visible phonon overtones \[14\] in the MIR spectrum led many authors in the past to a (bi)polaronic interpretation of the MIR \[13,15\]. In this model, the MIR absorption results from non-adiabatic carrier hopping processes where a different number of phonons is emitted and reabsorbed, the difference in energy being taken up by the IR photon. However, the measured MIR spectral shape shows significant deviations from the predicted polaronic spectrum \[18,50\] and expansion of this simple polaron hopping model to include a realistic density of phonon states and allowing for phonon dispersion still cannot reproduce the highly asymmetric peak in the MIR data. However, when we consider a two-component state, an additional process becomes possible, namely the excitation of carriers from the polaronic bound state to the unbound Fermionic state \[10\]. The expected MIR spectrum in this case exhibits an asymmetric spectral shape much closer to the experimentally observed one. A more sophisticated attempt at describing the MIR lineshape in detail might include both excitations from the localized states to the delocalized band \[19\] as well as non-adiabatic \[20\] hopping processes between localized states.

The interpretation of the MIR given above is supported by recent resonant electronic Raman spectra of Ruani and Ricci \[21\], where an electronic peak in the region of 0.06 eV (500 cm\(^{-1}\)) is observed below \( T^* \). The authors attribute this peak to excitations between dispersionless polaronic bands near the S-point of the Brillouin zone. Further support for the proposed interpretation comes from the observation of a depression in the density of states below \( T^* \) in the same region of the Brillouin zone in ARPES \[12\] to satisfy the sum rule, (bi)polaron formation (i.e. pairing) below \( T^* \) must be accompanied by a depletion in the density of states near \( E_F \), causing the appearance of a pseudo-gap in the ARPES spectrum.

The present analysis is essentially an alternative phenomenological view of the electrodynamic response of the cuprate superconductors to the one-component memory function parametrization more usually discussed in the recent past \[22,23\]. We believe it provides additional physical insight into the normal and superconducting state properties in line with the numerous recent suggestions for the co-existence of two structural local units in these materials \[10\]. Analysis of the optical conductivity in YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) as a function of \( \delta \) and \( T \) reveals some seemingly universal features which could be considered as a possible definition of the low-energy charge excitation spectrum for the cuprates:

1. The \( a-b \) optical conductivity \( \sigma_{\text{ab}} \) or \( \sigma_{\text{ab}} \) (with or without chains respectively) in YBCO universally displays a break from monotonic behaviour near 400 cm\(^{-1}\) (50 meV) below some temperature \( T^* \). The changes in the optical spectrum below \( T^* \) suggest a separation of \( \sigma_{\text{ab}} \) into two separate responses with characteristic excitation energy scales and well-defined and different \( \omega \) and \( T \)-dependences, rather than the opening of a simple gap. Above \( T^* \) the two excitations appear indistinguishable, presumably as a result of thermal fluctuations.

2. Below \( T^* \) the low-frequency optical conductivity (\( \omega < 400 \text{ cm}^{-1} \)) of the itinerant carriers is apparently described reasonably well by an extended Drude model with a frequency- and temperature-dependent scattering rate \( \tau_s \) \[20\]. Assuming a Fermi energy of 0.1 eV and \( m^* = 4m_e \), we find that the mean free path \( \lambda_c = v_F \tau_s \) for the Drude carriers at \( T^* \) is \( \sim 25 \) Å which is remarkably close to the typical length scale of the spatial inhomogeneities (stripe widths) found in structural experiments \[1,13\], suggesting that the properties of the Drude carriers are determined by the physics of the Fermions in the confined structure. The increasing \( \lambda_c \) at lower temperatures might be a sign of increasing coherence between stripes, rather than increasing stripe width.

3. The 2-component analysis is consistent both with the observations of a "pseudogap" in transport \[20\], NMR \[18\] and ARPES \[12\]. An explanation consistent with all four experimental techniques involves the formation of polarons in carrier-poor regions below \( T^* \) giving rise to a reduction in the DOS at \( E_F \) and hence a drop in the \( \chi\)(\( \omega = 0 \)) for NMR, as well as an apparent shift in the binding energy \( E_b \) in ARPES. This polaronic effect is indeed expected to be associated with the flat, dispersionless regions of the Fermi surface, consistent with the enhanced polaron effective mass \( m^* \), the electronic Raman data \[51\] and ARPES \[12\].

4. The in-plane MIR conductivity \( \sigma_{\text{ab}}^{\text{MIR}} \) which is presumed to be principally due to the excitation of localized carriers to the Fermionic regions in the Cu-O planes - increases in intensity systematically with \( \delta \) closely following the density of doped holes and is only weakly temperature dependent.
5. Numerous phonon anomalies appear in the optical conductivity in a range of temperatures such that $T_{anomaly} < T^*$, suggesting a coupling of the lattice degrees of freedom to the electronic charge excitations. Unfortunately there is insufficient data available at present for a more detailed examination of which phonons are involved, but the structural data from EXAFS and PDF appear to corroborate this observation. The $T^*$ observable for charge excitations appears to be consistent with $T_{sg}$ in NMR and other experimental techniques, and is at this time experimentally indistinguishable from it.

Finally, we briefly discuss the possible involvement of (bi)polarons in superconductivity. Their role was usually dismissed as unimportant on the grounds that there is no observable change in $\sigma(\omega)$ at MIR frequencies at $T_c$. In the BCS scenario, pairing occurs simultaneously with the opening of a gap in the charge excitation spectrum and the appearance of a macroscopic phase coherent state at (or very near) $T_c$. As a result of the gap, a significant change in spectral weight is observed in $\sigma(\omega)$ below $T_c$. In contrast, in the BE condensation picture, pairing is an independent process from condensation and the establishment of phase coherence. Of course it must occur above (or at) the condensation temperature because the process of condensation requires a macroscopic number of particles with Bose statistics to be present. In the case of underdoped cuprates, where neither the Drude nor MIR show any change at $T_c$ itself, the implication is that BE condensation of pre-formed pairs takes place with no change in pairing amplitude. It is natural to presume that because the redistribution of spectral weight in the optical spectrum starts to occur at $T^*$, this temperature should be associated with the onset of pair formation. Hence we conclude the paper by pointing out that the suggested two component interpretation of the optical conductivity spectra in underdoped cuprates is consistent with pair formation at $T^*$ and the Bose-Einstein condensation of such pairs across, or together with itinerant 'Drude' carriers into a macroscopically coherent ground state at $T_c$.

I. ACKNOWLEDGMENTS

The work was partly supported by the EU. One of us (D.M.) wishes to acknowledge discussion with D.Van der Marel and V.V.Kabanov.

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Figure 2. $T^*$ as a function of $T_c$ as determined from the appearance of an inflexion point in the in-plane optical conductivity. Seemingly there is no significant difference between $\sigma_{iab}$ and $\sigma_{1a}$. Insert: $T_{anomaly}$ as a function of $T_c$ as determined from phonon anomalies in the infrared and Raman spectra of 123, 124 and 247. Full symbols represent Raman data.

Figure 3. The phase diagram of YBCO determined from the in-plane optical conductivity. Seemingly there is no significant difference between $\sigma_{iab}$ and $\sigma_{1a}$.

Figure 4. The magnitude of the "residual" MIR as a function of a) O content and b) $T_c$ in YBaCuO. The upper data are for $\sigma_{iab}$, while the lower data set are for $\sigma_{1a}$. 
D. Mihailovic et al.: FIGURE 2

![Graph showing the relationship between $T_\text{c}$ and $T^*$ with data points from various references.](image-url)

- **$T_\text{c}$ (K)** on the x-axis.
- **$T^*$ (K)** on the y-axis.

Data points are labeled with references:
- ref. 28
- ref. 31
- ref. 32
- ref. 33
- ref. 34
- ref. 35
- ref. 36
- ref. 37
- ref. 38
- ref. 39
- ref. 40
- ref. 41
- ref. 42

The graph includes an inset with a higher density of data points, indicating a concentration of data at lower $T_\text{c}$ values.
D. Mihailovic et al.: Figure 3

The graph shows a plot of temperature (K) against oxygen content for two different phases, labeled as $T^*$ and $T_c$. The data points are represented by open circles and filled squares, with error bars indicating the uncertainty in the measurements. The graph illustrates the relationship between temperature and oxygen content, with $T^*$ and $T_c$ marking significant transitions or critical points in the material's properties.
D. Mihailovic et al.: FIGURE 4

MIR peak intensity (at 1000 cm\(^{-1}\)) ((Ω cm\(^{-1}\))

\[ \sigma_{1a} \]

Oxygen content

\[ T_c (K) \]

\[ \sigma_{1ab} \]

\[ \sigma_{1a} \]

- ■ ref. 28
- ▲ ref. 40
- ● ref. 41
- ▼ ref. 42