Correlation between the Josephson coupling energy and the condensation energy in bilayer cuprate superconductors

Dominik Munzar\textsuperscript{1,2}, Christian Bernhard\textsuperscript{2}, Todd Holden\textsuperscript{2}, Andrzej Golnik\textsuperscript{2,\ast}, Josef Humlíček\textsuperscript{1}, Manuel Cardona\textsuperscript{2}

\textsuperscript{1) Department of Solid State Physics, Faculty of Science, Masaryk University, Kotlářská 2, CZ-61137 Brno, Czech Republic
\textsuperscript{2) Max-Planck-Institut für Festkörperforschung, Heisenbergstraße 1, D-70569 Stuttgart, Germany

We review some previous studies concerning the intra-bilayer Josephson plasmons and present new ellipsometric data of the \(c\)-axis infrared response of almost optimally doped Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_8\). The \(c\)-axis conductivity of this compound exhibits the same kind of anomalies as that of underdoped YBa\(_2\)Cu\(_3\)O\(_7\)\(_{-\delta}\). We analyze these anomalies in detail and show that they can be explained within a model involving the intra-bilayer Josephson effect and variations of the electric field inside the unit cell. The Josephson coupling energies of different bilayer compounds obtained from the optical data are compared with the condensation energies and it is shown that there is a reasonable agreement between the values of the two quantities. We argue that the Josephson coupling energy, as determined by the frequency of the intra-bilayer Josephson plasmon, represents a reasonable estimate of the change of the effective \(c\)-axis kinetic energy upon entering the superconducting state. It is further explained that this is not the case for the estimate based on the use of the simplest “tight-binding” sum rule. We discuss possible interpretations of the remarkable agreement between the Josephson coupling energies and the condensation energies. The most plausible interpretation is that the interlayer tunneling of the Cooper pairs provides the dominant contribution to the condensation energy of the bilayer compounds; in other words that the condensation energy of these compounds can be accounted for by the interlayer tunneling theory. We suggest an extension of this theory, which may also explain the high values of \(T_c\) in the single layer compounds Tl\(_2\)Ba\(_2\)CuO\(_6\) and HgBa\(_2\)CuO\(_4\), and we make several experimentally verifiable predictions.

I. INTRODUCTION

The interlayer tunneling (ILT) theory\cite{1,2,3} provides a simple explanation of the surprisingly high values of \(T_c\) in the cuprate superconductors. It is based on the idea\cite{1,2,3,4} that the pairing mechanism is substantially amplified by a decrease of the effective \(c\)-axis kinetic energy of the electrons upon entering the superconducting state. A prerequisite for this decrease is the absence (or at least a strong suppression) of the coherent single particle tunneling between the copper-oxygen planes. The onset of the Cooper pair tunneling by the Josephson mechanism at \(T_c\) then leads to the decrease of the kinetic energy. According to the ILT theory, this gain of energy, which can be expressed\cite{5} as the negatively taken coupling energy of the internal Josephson junctions \((E_J)\), represents the dominant part of the condensation energy of the superconductor \((U_0)\),

\[ E_J \approx U_0. \]  

(1)

This relation, which should be exact in the limit of negligible in-plane contribution to the condensation energy and negligible coherent single particle tunneling, has been shown to be only moderately violated for La\(_{2-x}\)Sr\(_x\)CuO\(_4\)\cite{6} but strongly violated for Tl\(_2\)Ba\(_2\)CuO\(_6\) (Tl-2201)\cite{7,8,9} and for HgBa\(_2\)CuO\(_4\) (Hg-1201)\cite{10}. Here we show that Eq. (1) is fulfilled for two compounds that have two copper-oxygen planes per unit cell (bilayer compounds): YBa\(_2\)Cu\(_3\)O\(_7\)\(_{-\delta}\) (Y123) and Bi\(_2\)Sr\(_2\)CaCu\(_2\)O\(_8\) (Bi-2212).

So far the relation (1) has been tested only for single layer compounds, since it was not clear what kind of coupling takes place for the closely spaced copper-oxygen planes of the bilayer compounds. Anderson assumed that even these planes are only weakly coupled and argued that the bilayer compounds consist of two kinds of Josephson junctions: inter-bilayer and intra-bilayer\cite{11}. The Josephson plasma frequency of the inter-bilayer junction \((\omega_{\text{int}})\) is lower than that of the intra-bilayer junction \((\omega_{\text{in}})\). The former determines the \(c\)-axis penetration depth while the latter
determines $U_0$. Van der Marel and Tsvetkov [12] proposed a phenomenological model of the dielectric response of such a superlattice of inter-bilayer and intra-bilayer Josephson junctions. They showed that it exhibits a transverse resonance between the two zero crossings corresponding to the two plasmons. It has been suggested [13] that this new excitation (“transverse plasma excitation”), which can be visualized as a resonant oscillation of the condensate density between the two closely spaced copper-oxygen planes, has indeed been observed as an additional absorption peak which appears at low temperature in the spectra of the infrared $c$-axis conductivity of underdoped Y123 [14-17]. Very recently, this interpretation has been put on a firm basis by a detailed analysis of the $c$-axis conductivity data for Y123 with different oxygen concentrations [13,4]. Note that the observation of the transverse plasma excitation also implies the existence of the second (intra-bilayer) Josephson plasmon which is a vital ingredient of the ILT theory. It opens a possibility to test also for bilayer compounds the relationship between the Josephson coupling energy and the condensation energy predicted by the ILT theory (Eq. (1)).

In this paper we review some of the previous experimental observations of the transverse plasma excitation (TPE). In addition, new data for almost optimally doped Bi-2212 with $T_c \approx 91$ K are reported and analyzed (section II). In section III, the Josephson coupling energies of different bilayer compounds obtained from the $c$-axis conductivity data of Refs. [14,15,16,17] and of the present work are compared with the condensation energies obtained from the specific heat data [22,23]. Section IV contains a discussion of the frequently used sum-rules-based estimates of the changes of the $c$-axis kinetic energy [24,25]. Finally, an extension of the interlayer tunneling theory is proposed in section V, which allows one to explain the high values of $T_c$ in the single layer compounds Tl-2201 and Hg-1201.

II. RESULTS AND DISCUSSION

Since the early days of the high-temperature superconductivity, it was known that some of the infrared active $c$-axis phonon modes of the cuprate superconductors exhibit changes (so-called “phonon anomalies”) in the vicinity of the superconducting transition temperature (see, e.g. Refs. [27,28,29]). The most pronounced anomalies have been observed for underdoped Y123 [14,15,16,17].

A. YBa$_2$Cu$_3$O$_{7-\delta}$

The anomalies are illustrated in Fig. 1, which shows our experimental spectra of the $c$-axis conductivity of YBa$_2$Cu$_3$O$_{6.55}$ with $T_c = 53$ K [18]. Note the following anomalies. (A) As the temperature decreases, the oxygen bond-bending mode at 320 cm$^{-1}$ which involves the in-phase vibration of the planar oxygens against the Y-ion and the chain ions [21,22] loses most of its spectral weight and softens by almost 20 cm$^{-1}$; (B) at the same time a new broad absorption peak appears in the spectra around 410 cm$^{-1}$ and (C) the spectral weight of the peak corresponding to the apical oxygen mode at 560 cm$^{-1}$ decreases. The additional absorption peak (feature (B)) has been sometimes considered to be a new phonon [15]. On the other hand, van der Marel and coworkers suggested [13] that it corresponds to the TPE of their model [12]. We believe that this interpretation is correct, but one has to keep in mind that some part of the spectral weight of the peak ($\sim 50\%$) indeed comes from the phonons (see Ref. [17] for a detailed discussion). With increasing doping the additional peak shifts towards higher frequencies and it becomes broader and less pronounced [15,17]. Although the anomalies start to develop above $T_c$ in strongly underdoped compounds, the pronounced and steep changes always occur right at $T_c$ [17]. Similar though less spectacular effects have also been observed for other values of $\delta$ [14,15,16,17,18,19,20].

B. Bi$_2$Sr$_2$CaCu$_2$O$_8$

Very recently, Železný et al. [21,22] have reported similar anomalies in the infrared $c$-axis conductivity of underdoped Bi-2212. The spectra exhibit an increase of the electronic background around 450 cm$^{-1}$ below $T_c$ accompanied by characteristic phonon anomalies. The increase of the background corresponds to the additional absorption peak such as observed in the spectra of underdoped Y123. The most pronounced phonon anomaly again consists in a sizeable decrease of the spectral weight of the 355 cm$^{-1}$ phonon mode, which has —according to the shell model calculations
— a similar eigenvector as the 320 cm\(^{-1}\) mode in Y123. The doping and temperature dependence of the anomalies is close to that observed in Y123. These findings of Železný \textit{et al.} confirmed that the anomalies are a common property of the bilayer cuprate compounds.

Fig. 2 shows our experimental spectra of the c-axis optical conductivity of an almost optimally doped Bi-2212 single crystal with \(T_c = 91\) K which have been obtained by ellipsometric measurements (see Ref. [17] and references therein for a description of the technique). The measurements have been performed on a large \(ac\)-face of size \(4 \times 0.5\) mm\(^2\). As shown in Fig. 2 (b), the spectra exhibit the same kind of anomalies as underdoped Y123 [4,15,16,17,18] and underdoped Bi-2212 [19,20]. Below \(T_c\) the electronic background increases in the frequency region around 500 cm\(^{-1}\) (feature (B)) and simultaneously some of the infrared-active phonon modes are renormalized; in particular, the phonon mode at 355 cm\(^{-1}\) loses a large part of its spectral weight (feature (A)). As compared to the data for underdoped Bi-2212, the region of the spectral weight increase is shifted towards slightly higher frequencies. This can be expected because the frequency of the TPE is known to increase with increasing hole doping (see Ref. [18] for a discussion). The onset temperature of the anomalies coincides with \(T_c\) (cf. the insets of Fig. 2 (a) and Fig. 2 (b)) which confirms that they are related to superconductivity. Note that some signatures of the anomalies can also be found in the reflectivity data of Ref. [21].

Figure 3 (a) displays the region near the anomalies on an enlarged scale and Fig. 3 (b) shows the fits obtained by using a slightly modified version [21] of the model of Ref. [18]. The modification concerns only the description of the featureless electronic background. First, the inter-bilayer susceptibility is set equal to zero \((\chi_{\text{int}} = 0\) in notation of Ref. [18]). This is a reasonable approximation considering the very low value of the unscreened inter-bilayer plasma frequency \(\omega_{\text{int}}\) \((\omega_{\text{int}} \leq 20\) cm\(^{-1}\), see Ref. [13]). Second, the regular part of the intra-bilayer susceptibility \(\chi_{bl}\) has been fitted by a combination of a broad Drude term and a broad mid-infrared Lorentz oscillator instead of the single very broad Lorentzian centered at high frequencies which was used previously. Furthermore, we have adopted the simple picture of the phonon eigenvectors used in the fits of the spectra of underdoped Y123 and underdoped Bi-2212, i.e., we assume that the 355 cm\(^{-1}\) mode corresponds to the vibrations of the planar oxygens, whereas the modes at 300 cm\(^{-1}\) and 580 cm\(^{-1}\) correspond to vibrations of the inter-bilayer oxygens, i.e., the apical oxygens and the oxygens of the BiO layers. The values of the fitting parameters and other numerical factors used are summarized in Table I. Those corresponding to the room-temperature spectrum have been obtained by fitting the measured frequency dependence of the complex dielectric function in the frequency region above 300 cm\(^{-1}\) (with \(\omega_{bl} = 0\) and \(S_{bl} = 0\), \(S_{bl}\) being the oscillator strength of the mid-infrared Lorentz oscillator). Those values obtained in calculating the 100 K spectrum have also been obtained by fitting the data (with \(\omega_{bl} = 0\) and \(S_{bl} = 0\), except for the values of \(\varepsilon_{\infty}, S_P, S_1, S_2\) (oscillator strengths of the phonons), \(\omega_P\) (frequency of the oxygen-bond bending mode), \(\omega_1\) and \(\gamma_1\) (parameters of the 300 cm\(^{-1}\) mode), which have been fixed at the room-temperature ones. Finally, the values of \(\varepsilon_{\infty}, S_P, S_1, S_2, \omega_P, \gamma_P, \omega_1, \gamma_1, \) and \(\gamma_{bl}\) (broadening parameter of the broad Drude contribution), have been fixed at the 100 K ones, when fitting the low-temperature spectrum. It can be seen that the most pronounced features of the experimental spectra, in particular the increase of the electronic background around 550 cm\(^{-1}\) and the spectral-weight anomaly of the oxygen bond-bending mode, are well reproduced.

Figure 3 (c) shows the frequency dependence of the difference \(\sigma_1(T = 10\) K \(<< T_c) - \sigma_1(T = 100\) K \(\approx T_c)\). The value of the difference is positive in the frequency region between 420 cm\(^{-1}\) and 580 cm\(^{-1}\), whereas it is negative both for lower frequencies (in the region between 340 cm\(^{-1}\) and 420 cm\(^{-1}\)) and for higher frequencies (in the region between 580 cm\(^{-1}\) and 650 cm\(^{-1}\)). The positive values in the first region are caused mainly by the increase of the electronic background below \(T_c\) (i.e., the additional peak due to the TPE). The negative values in the second region correspond to the spectral weight anomaly of the oxygen bond-bending mode. The negative values in the third region are mainly due to a slight shift of the apical-oxygen mode at 580 cm\(^{-1}\) towards lower frequencies. The agreement between the difference of the measured data (full line) and the difference of the fitted spectra (dashed line) is excellent. Note, that this agreement has not been achieved by changing the values of the parameters of the phonon modes. The slight shift of the 580 cm\(^{-1}\) mode accounts only for a part of the spectral weight increase around 550 cm\(^{-1}\).

Finally, the frequency dependence of the quantity \(N_n(N_s) - N_s\), where \(N_n(\omega) = N(T = 100\) K \(\approx T_c, \omega)\), \(N_s(\omega) = N(T = 10\) K \(<< T_c, \omega)\) and

\[
N(T, \omega) = \int_0^\omega d\omega' \sigma_1(T, \omega'),
\]

is displayed in Fig. 3 (d). We take our low-frequency cutoff of 70 cm\(^{-1}\) as the lower limit of the integration in Eq. (2), but it is rather unlikely that below this frequency there is any considerable difference between the normal state and the superconducting state data. For a conventional superconductor, the value of \(N_n - N_s\) increases with increasing frequency and it approaches \(\rho_s\), the spectral weight of the \(\delta\)-peak at \(\omega = 0\), within a range of \(\sim 6\Delta\), \(\Delta\) being the superconducting gap [13]. It has been shown by Basov \textit{et al.} [27] that for several high-temperature superconductors the value of \(N_n - N_s\) also increases with increasing frequency but it saturates at a value of only about one half of
\(\rho_s\) for \(\omega \sim 10\Delta\). Our result for Bi-2212 exhibits an even more surprising tendency: \(N_n - N_s\) stays approximately constant up to \(\sim 350\ \text{cm}^{-1}\), it changes considerably in the frequency region between \(350\ \text{cm}^{-1}\) and \(620\ \text{cm}^{-1}\) and it seems to saturate above \(\sim 650\ \text{cm}^{-1}\) at a negative value of approximately \(-300\ \Omega^{-1}\ \text{cm}^{-2}\). The corresponding value of the ratio \(|N_n - N_s|/\rho_s\) is larger than 25. The fact that the value of \(N_n - N_s\) above \(\sim 550\ \text{cm}^{-1}\) is negative signals an increase of the low frequency spectral weight below \(T_c\), which is caused by the formation of the peak due to the TPE. Note that such an increase cannot be easily explained by any conventional theory. We shall come back to this interesting issue in section IV B.

### III. JOSEPHSON COUPLING ENERGIES AND CONDENSATION ENERGIES

In Table II we summarize the values of the intra-bilayer plasma frequency \((\omega_{bl})\), the corresponding Josephson coupling energy per unit cell \((E_J)\), and the condensation energy per unit cell \((U_0)\) for Y123 and Bi-2212. The values of \(E_J\) have been calculated using the formula [16]

\[
E_J = \frac{\hbar^2 \varepsilon_0 a^2 \omega_{bl}^2}{4e^2 d_{bl}} \quad \text{or equivalently} \quad E_J[\text{meV}] = \frac{C}{d_{bl}[\text{Å}]} (\omega_{bl}[\text{cm}^{-1}])^2.
\]

Here \(a\) is the in-plane lattice constant, \(d_{bl}\) is the distance between the closely spaced copper-oxygen planes, and \(C = 3.1 \times 10^{-7}\). We neglect the contribution of the inter-bilayer plasmon to \(E_J\) which is by at least one order of magnitude lower than that of the intra-bilayer one. Note that Eq. (3) does not contain any adjustable parameters. The values of the condensation energies have been obtained from the specific heat data in Ref. [22] (Y123) and in Ref. [23] (Bi-2212). We estimate the error bars of the values of \(\omega_{bl}\) to be about 20 – 30\%. These error bars arise mainly from the uncertainty in the description of the electronic background [17]. The corresponding uncertainties of the values of \(E_J\) are about 50\%. Taking this into account, the agreement between the Josephson coupling energies obtained from the optical data and the condensation energies obtained from the specific heat data is reasonably good:

(i) The values of the two quantities are of the same order of magnitude.

(ii) Both quantities exhibit a rather similar dependence on doping. It is even possible to understand, why the values of the ratio \(E_J/U_0\) are higher for the strongly underdoped samples than for the less underdoped ones or the optimally doped ones. It is namely likely that for strongly underdoped samples some fluctuation effects set in well above the macroscopic transition temperature \(T_c\). This suggestion is consistent with the finding that the phonon anomalies start to occur well above \(T_c\) in strongly underdoped samples [17,18]. The contribution of the fluctuation effects to the condensation energy is not likely to be contained in the values of \(U_0\) presented in Table II, since they have been obtained by an analysis of the specific heat data where only the changes occurring below \(T_c\) are properly taken into account. On the other hand, the values of \(E_J\) do contain the contribution of these effects, since they are determined by the low-temperature values of \(\omega_{bl}\) (i.e., in a way which does not require any assumptions concerning the onset of superconductivity). As a result the values of \(U_0\) for strongly underdoped samples can be expected to be smaller than the values of \(E_J\).

(iii) The values of \(E_J\) for Y123 are systematically higher than those for Bi-2212, in agreement with the trend in the condensation energies. It may be argued that the difference in the condensation energies is related to the presence of metallic chains in Y123. However, the chain condensation cannot fully account for the difference, since Ca substituted samples with broken chains also have a significantly higher condensation energy than Bi-2212 [2].

This agreement means that the values of the condensation energies of the bilayer compounds can be explained using the ILT theory. Further implications will be discussed in Section V. It certainly remains an open question why different compounds with almost identical values of \(T_c\) have rather different values of the condensation energy.

### IV. SUM RULES

There is another approach to estimate the changes of the \(c\)-axis kinetic energy, pioneered by Chakravarty, Kee and Abrahams [4][24], which is based on the use of the optical sum rule. For the bilayer compounds, this approach yields an opposite result, namely that the value of the condensation energy cannot be accounted for by the change of the \(c\)-axis kinetic energy. In the first part of this section we clarify several conceptual points which have made the discussions of the kinetic energy changes somewhat confusing (subsection A). Next we discuss some specific properties of the bilayer compounds (subsection B). In particular, we show that for these compounds the change of the \(c\)-axis kinetic energy cannot be estimated using the conventional “tight-binding” sum rule of Refs. [124].
A. Conceptual issues

For simplicity, we focus in our considerations first on the single layer compounds. In the present context, there are at least three different quantities denoted as c-axis kinetic energy. First, the “true” c-axis kinetic energy, $K_{c} = \sum_{l} p_{l,c}^{2}/2m$. Second, the “tight-binding” c-axis kinetic energy $H_{c}$,

$$H_{c} = -\sum_{l,i,j,s} t_{\perp}(l,i,j)c_{j,l+1,s}c_{i,l,s} + H.c.,$$

where $l$ is the layer index, $i$ and $j$ refer to the sites of the two-dimensional layers, $s$ refers to spin, $t_{\perp}$ is the interlayer hopping matrix element, and $H.c.$ means the Hermitian conjugate operator. Third, the effective low-energy c-axis kinetic energy of the ILT theory $H_{f}$,

$$H_{f} = -\sum_{l,k} T_{j}(k)c_{k,l+1,↑}c_{-k,l+1,↑} - k,l,↓c_{k,l,↑} + H.c.,$$

i.e., the Josephson coupling energy. The experimental data are discussed in terms of at least two different sum rules. First, the “general” sum rule (for derivation see, e.g., Ref. [49]),

$$\int_{0}^{\infty} \sigma_{1}(T,\omega) d\omega = \frac{\pi n e^{2}}{2m},$$

which is fulfilled for any real system and second, the “tight-binding” sum rule valid only for model Hamiltonians whose single-particle part is of the tight-binding form (for derivation see, e.g., Refs. [50, 4] and references therein),

$$\int_{0}^{\infty} \sigma_{1}(T,\omega) d\omega = -\frac{\pi e^{2}d}{2\hbar^{2}a^{2}}(H_{c})(T).$$

In Eq. (6), $n$ is the total electron density, in Eq. (7) $a$ is the in plane lattice constant, $d$ is the lattice constant along the c-axis, and $H_{c}$ is the tight-binding kinetic energy (Eq. (4)) per unit cell ($\langle H_{c} \rangle$ represents an average of this quantity, $T$ is the temperature).

Let us discuss possible changes of the three kinetic energies upon entering the superconducting state and the relations between them and the sum rules.

(i) There is no obvious relation between $\langle K_{c} \rangle$ and the sum rules. Upon entering the superconducting state, the total energy of the superconductor decreases. This implies—by virtue of the virial theorem ($\langle K \rangle = -\langle V \rangle/2$, where $K$ and $V$ are the kinetic and the potential energy, respectively; see, e.g., Ref. [3])—that its total kinetic energy increases. It is possible to assume that even $\langle K_{c} \rangle$ increases at the superconducting transition.

(ii) It is very likely that the degrees of freedom essential for superconductivity are contained in some model single band Hamiltonian $H_{b}$ whose (tight-binding like) single particle part is derived by a downfolding process in which all of the higher energy bands are integrated out and whose interaction part describes the most pronounced correlation effects. In our opinion, there are no general reasons why the average of its c-axis kinetic energy term $H_{c}$ (Eq. (4)) should decrease or increase below $T_{c}$. The arguments invoking a frustration of the c-axis kinetic energy in the normal state and a “deconfinement” of bosonic pairs below $T_{c}$ (See, e.g., Ref. [3], Chapter 2, Dogma VI and Ref. [3]) are usually based on asymptotic properties of the quasiparticle propagators, i.e., they concern rather the asymptotic behaviour for $\omega \rightarrow 0$ than the actual changes of $\langle H_{c} \rangle$. Possibly these arguments could be formulated more precisely in terms of yet another effective kinetic energy $H'_{c}$ characterising the c-axis electrodynamics in a frequency scale intermediate between that of $H_{c}$ ($\sim 10000$ cm$^{-1}$) and the frequency scale of $H_{f}$ of the order of $100$ cm$^{-1}$ (see Fig. 4).

It follows from Eq. (7) that the possible change of $\langle H_{c} \rangle$ at the superconducting transition is related to the change of the spectral weight as follows:

$$\langle H_{c} \rangle_{s}(T << T_{c}) - \langle H_{c} \rangle_{n}(T \approx T_{c}) [\text{meV}] = -\frac{4C}{d[A]} \frac{120}{\pi} \left( \int_{0}^{\Omega_{c}} \sigma_{1}(T << T_{c}, \omega) d\omega - \int_{0}^{\Omega_{c}} \sigma_{1}(T \approx T_{c}, \omega) d\omega \right) [\Omega^{-1} \text{cm}^{-2}],$$

where $\Omega_{c}$ is a cutoff frequency required to exhaust the sum rule (7). Note that in a model involving strong correlations, the value of $\Omega_{c}$ may be much higher than the bandwidth of the corresponding noninteracting model. In addition, this
value (and also the changes of the kinetic energy themselves) may depend on the kind of model which is used, i.e., on the interaction part of the model Hamiltonian $H_{1b}$.

(iii) We emphasize that it is the effective c-axis kinetic energy $H_J$ of the ILT theory (Eq. (5)) whose changes at the superconducting transition were predicted to be responsible for the high values of $T_c$ in Ref. [1]. The only “sum rule”, which can be associated with $H_J$, is Eq. (3) with $d\omega/2\pi$ substituted by $d$ (note that $E_J = -(H_J)$).

What is the experimental status? The analysis of Basov and coworkers [25] demonstrates that at least for the underdoped high-$T_c$ superconductors, the low-frequency spectral weight

$$\alpha(T,\omega) = \int_0^\omega \sigma_1(T,\omega') d\omega' = \rho_s(T) + N(T,\omega)$$  \hspace{1cm} (9)

increases considerably upon entering the superconducting state for cutoff frequencies $\omega$ ranging up to at least 1200 cm$^{-1}$. More precisely, as one increases the value of $\omega$, the value of the difference $\alpha(T << T_c,\omega) - \alpha(T \approx T_c,\omega)$ saturates at $\omega \approx 500$ cm$^{-1}$, reaching typically 50% of $\rho_s$. The only way to reconcile this interesting finding with the general sum rule (6) consists in assuming [25] that a substantial part of $\alpha(T << T_c, 1200$ cm$^{-1}$) is collected from frequencies exceeding 1200 cm$^{-1}$. In other words, since the total spectral weight has to be conserved, the increase of $\alpha(T,1200$ cm$^{-1}$) below $T_c$ is compensated by a decrease of spectral weight around some frequency $\omega^*$ higher than 1200 cm$^{-1}$. A sketch of the situation is shown in Fig. 4.

What do we learn from these results about the changes of the tight-binding kinetic energy $\langle H_c \rangle$? Let us denote by $\omega_c$ the interband cutoff, i.e., the upper limit of the frequency interval for which the response of the superconductor can be properly described using a certain effective Hamiltonian $H_{1b}$ (e.g., the Hubbard Hamiltonian). Let us further assume, for the sake of simplicity, that $\omega_c \geq \Omega_c$ of Eq. (8) [54]. There are two different possibilities, which are both compatible with the infrared data: (a) $\omega^* > \omega_c$ and (b) $\omega^* < \omega_c$ (see Fig. 4). In case (a), it follows from Eqs. (6) and (8) that $\langle H_c \rangle$ indeed decreases below $T_c$. In order to establish this decrease experimentally, however, one would have to estimate the total low-frequency spectral weight above and below $T_c$, i.e., to integrate $\sigma_1(\omega)$ up to $\omega_c$. We believe that the case (b) is more likely to happen. The increase of $\alpha(T,1200$ cm$^{-1}$) below $T_c$ and the corresponding decrease of the spectral weight around $\omega^*$ then both can be described by using $H_{1b}$. It follows from Eqs. (6) and (8) that $\langle H_c \rangle$ does not change at $T_c$. To conclude, it is only the tight-binding c-axis kinetic energy $H_c$ (Eq. (4)) of an effective single band Hamiltonian, whose change at the superconducting transition can be obtained using Eq. (8). The results of Basov et al. [25] nicely reveal the unconventional properties of the high-temperature superconductors, in particular the extremely large frequency scale involved, but they cannot be used to yield reliable estimates of this change because of the uncertainties concerning the cutoff frequencies $\Omega_c$ and $\omega_c$. In addition, there are no apriori reasons, why $\langle H_c \rangle$ should change upon entering the superconducting state.

Another example of a model c-axis kinetic energy is represented by the effective low-energy c-axis kinetic energy $H_J$ (Eq. (5)) of Ref. [1]. It acquires a nonzero (negative) value, $-E_J$ (Eq. (3)), only in the superconducting state and it was predicted to be responsible for the high values of $T_c$. It is certainly a crude approximation to identify $E_J$ with the c-axis contribution to the condensation energy, as we did it in the previous section, since possible “countereffects” [55] are not included. They are to some extent included when estimating the changes of the c-axis kinetic energy using Eq. (8) with the upper limit of the integrals well below $\omega^*$ (instead of $\omega_c$), i.e.,

$$\Delta E_{\text{kin,c}}[^\text{meV}] = -\frac{4C}{d[A]} \frac{120}{\pi} \left(\alpha(T << T_c,\omega < \omega^*) - \alpha(T \approx T_c,\omega < \omega^*)\right) [\Omega^{-1} \text{ cm}^{-2}], \hspace{1cm} (10)$$

This is an appealing possibility, but the physical meaning of the result is not completely clear (in contrast to the change of the well defined quantity $\langle H_c \rangle$). It may be perhaps viewed as an estimate of the change of some kinetic energy $H'_c$ characterising the c-axis electrodynamics in an intermediate frequency scale (see our discussion above and Fig. 4). Note finally that the fact that $\Delta E_{\text{kin,c}}$ includes the countereffects does not necessarily imply that $|\Delta E_{\text{kin,c}}| < E_J$ because of the additional factor of 4 entering the sum-rules-based formulas [50]. On the contrary, for moderate countereffects, $|\Delta E_{\text{kin,c}}| > E_J$. For underdoped La$_{2-x}$Sr$_x$CuO$_4$, e.g., $|\Delta E_{\text{kin,c}}|$ is by a factor of ~2 larger than $E_J$ [56].

### B. Some specific properties of the bilayer compounds

In Fig. 3 (d), we have encountered a rather unusual situation. First, the value of the quantity $N(T,690$ cm$^{-1}$) defined by Eq. (2) increases with decreasing temperature. A phenomenon not observed for any of the single-layer compounds (see, e.g., Ref. [23]). Second, the increase of the integrated spectral weight $\alpha(T,690$ cm$^{-1}$) defined by Eq. (9) upon entering the superconducting state is lower by a factor of ~30 than the value required to yield a kinetic
energy change comparable to \( U_0 \) (when inserted into Eq. (10)). On the other hand, using simply the formula (3) for the Josephson coupling energy, we have obtained a value fairly close to \( U_0 \). Here we propose a qualitative explanation of these paradoxes.

Let us consider the superlattice of intra-bilayer and inter-bilayer Josephson junctions. The low frequency spectral weight \( \alpha(T << T_c, \omega > \omega_p) \), where \( \omega_p \) is the frequency of the TPE (Eq. (A.2)), contains both the contribution of the \( \delta \)-peak at \( \omega = 0 \), \( S_\delta \) (Eq. (A.3)), and the contribution of the TPE at \( \omega = \omega_p \), \( S_{pl} \) (Eq. (A.4)). The appearance of the second one below \( T_c \) explains the first paradox. It is easy to show that

\[
\alpha(T << T_c, \omega > \omega_p) = \frac{\pi}{2} \frac{d_{pl} \omega_p^2 + d_{int} \omega_{int}^2}{d_{pl} + d_{int}} \approx \frac{\pi}{2} \frac{d_{pl} \omega_p^2}{d_{pl} + d_{int}},
\]

(11)

where \( d_{int} \) is the distance between the bilayers. The Josephson coupling energy is approximately given by Eq. (3) (neglecting the contribution of the inter-bilayer Josephson junction), whereas the change of the \( c \)-axis kinetic energy estimated using Eqs. (10) and (11) is given as follows:

\[
\Delta E_{kin,c}[\text{meV}] = - \frac{4C}{(d_{pl} + d_{int})[\text{Å}]} \frac{d_{pl} \left( \omega_{pl}[\text{cm}^{-1}] \right)^2}{d_{pl} + d_{int}}.
\]

(12)

It can be seen that

\[
E_J >> |\Delta E_{kin,c}|.
\]

(13)

This is the explanation of the second paradox. The effects of the electronic background make the discrepancy even more pronounced. At the same time, it is obvious that within the simple model of the superlattice of intra-bilayer and inter-bilayer Josephson junctions, it is the Josephson coupling energy \( E_J \), rather than \( \Delta E_{kin,c} \), which represents the change of the \( c \)-axis kinetic energy upon entering the superconducting state. We conclude that for the bilayer compounds the change of the \( c \)-axis kinetic energy can be estimated using Eq. (3) but cannot be estimated using Eq. (10). We refer the reader to appendix B for a more rigorous discussion.

V. POSSIBLE EXTENSION OF THE ILT SCENARIO FOR SINGLE LAYER COMPOUNDS

In section III we have shown that for the bilayer cuprate compounds the Josephson-coupling energy such as estimated from the optical data can account for the condensation energy. In section IV B we have shown that this result is not invalidated by the fact that Eq. (10) yields a much lower estimate of the change of the \( c \)-axis kinetic energy upon entering the superconducting state than Eq. (3). The main reason of the discrepancy is that the tight-binding sum rule of Eq. (7) used to derive Eqs. (8) and (10) is only valid for such single layer compounds, for which the distribution of the total electric field is homogeneous. The results presented in Section III thus suggest that the high-temperature superconductivity in the bilayer compounds can be accounted for by the ILT theory. We are left with three possible explanations.

(a) The agreement between the Josephson-coupling energies and the condensation energies reported in section III represents a mere coincidence.

(b) The interlayer tunnelling indeed provides the dominant contribution to the condensation energy of the bilayer compounds. Another mechanism is responsible for the high values of \( T_c \) in the single layer compounds Tl-2201 and Hg-1201.

(c) A modified ILT theory may explain the high-temperature superconductivity both in the bilayer compounds and in the single layer compounds.

In our opinion, it is unlikely that case (a) is realized. It would mean either that the assignment of the additional absorption peak to the TPE is wrong or that the Josephson-coupling energy is completely (or almost completely) compensated by the countereffects. There are several important arguments supporting the present interpretation of the data. Let us mention two of them. First, the doping dependence. The frequency of the maximum of the additional absorption peak increases considerably with increasing doping. This is easy to explain within the present scenario (the squared frequency of the maximum should be proportional to the condensate density) but difficult to explain within theories where the additional peak is assigned to a pair breaking excitation. According to these theories the frequency of the maximum should be close to the pair breaking frequency observed in other experiments, which seems not to be the case for strongly underdoped samples. One could argue that the final state interactions may shift the peak towards lower frequencies. Second, both the frequency and the spectral weight of the additional peak are determined by a single parameter, \( \omega_{pl} \). In other theories, at least two parameters
are required to fit the data, e.g., $\Delta$ and $t_\perp$. Starting from our interpretation of the data, large countereffects would represent the only possibility to rule out the ILT mechanism from the role of the mechanism providing the dominant contribution to the condensation energy $U_0$. Note that the values of the Josephson-coupling energies presented in Table II are so high that even sizeable countereffects of the order of 80% would not invalidate our interpretation (see Ref. [56]). A possible analysis of the countereffects along the lines of appendix B is complicated by the fact that the normal-state data for very low temperatures, which should be compared to the superconducting-state data, rather than the data obtained for temperatures above $T_c$, are not available. If case (b) were realized, there would be two different mechanisms causing the high values of $T_c$. This is certainly not impossible. In the remaining part of this section we argue that even the possibility (c) should not be excluded. We propose an extension of the ILT theory, which may explain the high-$T_c$ values in the single layer compounds TI-2201 and Hg-1201. For concreteness we focus on the TI-2201 compound, for which some optical data are already available [15,16].

According to the ILT theory as formulated, e.g., in Ref. [1], the $c$-axis kinetic energy is frustrated in the normal state because of the spin-charge separation mechanism. The electrons or holes are composite objects that cannot escape from the copper-oxygen planes (so called “confinement”). For Cooper pairs this confinement is relaxed and consequently the $c$-axis kinetic energy decreases at the superconducting transition. To our best knowledge, it has been previously always assumed that this decrease is related to the onset of Josephson tunneling between the superconducting copper-oxygen planes. This is, however, only one possible mechanism for the decrease. We suggest that in TI-2201 the $c$-axis kinetic energy decreases via some delocalization of the Cooper pairs which involves the apical-oxygen orbitals. By the delocalization we mean the fact that the superconducting-state wave function acquires a larger contribution of the kinetic energy decreases via some delocalization of the Cooper pairs which involves the apical-oxygen orbitals. The conjecture can be experimentally tested in several ways. First we discuss the hypothesis. Not only the hopping between the copper-oxygen planes is assumed to be blocked in the normal state but also (at least to some extent) the hopping between the orbitals of the copper-oxygen planes and the apical-oxygen orbitals. The conjecture can be experimentally tested in several ways. First we discuss the hypothesis. Not only the hopping between the copper-oxygen planes is assumed to be blocked in the normal state but also (at least to some extent) the hopping between the orbitals of the copper-oxygen planes and the apical-oxygen orbitals. The conjecture can be experimentally tested in several ways. First we discuss the hypothesis. Not only the hopping between the copper-oxygen planes is assumed to be blocked in the normal state but also (at least to some extent) the hopping between the orbitals of the copper-oxygen planes and the apical-oxygen orbitals. The conjecture can be experimentally tested in several ways. First we discuss the hypothesis. Not only the hopping between the copper-oxygen planes is assumed to be blocked in the normal state but also (at least to some extent) the hopping between the orbitals of the copper-oxygen planes and the apical-oxygen orbitals. The conjecture can be experimentally tested in several ways. First we discuss the hypothesis. Not only the hopping between the copper-oxygen planes is assumed to be blocked in the normal state but also (at least to some extent) the hopping between the orbitals of the copper-oxygen planes and the apical-oxygen orbitals. The conjecture can be experimentally tested in several ways. First we discuss the hypothesis. Not only the hopping between the copper-oxygen planes is assumed to be blocked in the normal state but also (at least to some extent) the hopping between the orbitals of the copper-oxygen planes and the apical-oxygen orbitals. The conjecture can be experimentally tested in several ways. First we discuss the hypothesis. Not only the hopping between the copper-oxygen planes is assumed to be blocked in the normal state but also (at least to some extent) the hopping between the orbitals of the copper-oxygen planes and the apical-oxygen orbitals.
The increase of $\Omega_A$ may simulate the changes brought about by the superconducting transition: the more delocalized the ground state wavefunction, the more metallic the region A and the higher the value of $\Omega_A$. For the sake of simplicity, we do not consider the changes of $\sigma_B$ caused by the onset of superconductivity. The values of the parameters used are given in Table III.

It can be seen in Figs. 5 (b) and 5 (d) that the lowest frequency mode manifests itself as a step rather than as a Lorentzian peak, in agreement with the experimental data. Note that the step is getting less pronounced with increasing value of $\Omega_A$, i.e., with increasing metallicity of the region A. This may correlate with an apparent absence of the structure in the data for strongly overdoped Tl-2201 (See Fig. 2 of Ref. [62]). As seen in Figs. 5 (a) and (c), the phonon mode involving vibrations of the TiO-layer oxygens exhibits distinct changes with increasing value of $\Omega_A$, in particular its spectral weight increases [63]. This may correspond to the observed spectral weight anomaly of the 390 cm$^{-1}$ phonon mode (assuming that the assignment $A2$ is closer to reality than the assignment $A1$). Finally, the increase of $\Omega_A$ results in an increase of the electronic background around 1000 cm$^{-1}$ (concerning the absolute values see Ref. [66]). Such an increase should be observed in future experiments. Note that in the present simulations, the increase of $\Omega_A$ is intentionally relatively high so that the anomalies are clearly seen. Such an increase would correspond to a change of the $c$-axis kinetic energy much higher (by a factor of $\sim 4$, a rough estimate obtained by using Eq. (B.11)) than the condensation energy [69]. The actual changes of the electronic background to be observed experimentally therefore should be considerably smaller (e.g., by a factor of 4). Also the frequency range of the increase may be somewhat different. To summarize, our simulations demonstrate that the anomalous features (i) and (ii) can be explained within a model involving the variations of the electric field inside the unit cell, whereas they cannot be easily explained in a conventional way. The important point is that if the electric field indeed changes considerably inside the unit cell, the changes of the $c$-axis kinetic energy upon entering the superconducting state cannot be estimated simply from the value of the plasma frequency of the superconducting condensate or by using the modified tight-binding sum rule (10).

Let us compare the $c$-axis conductivity of Tl-2201 with that of another single layer compound with considerably lower $T_c$, Bi$_2$Sr$_2$CuO$_6$ (Bi-2201). The spectra for Bi-2201 exhibit five peaks corresponding to the $c$-axis infrared phonons (See Fig. 2 of Ref. [1]), similar results have been obtained by our group [8], i.e., there is one phonon peak more than in the spectra for Tl-2201. This difference finds a natural explanation within our extension of the ILT theory. Bi-2201 has a substantially lower condensation energy than Tl-2201 [8,7]. This suggests that in Bi-2201 the apical-oxygen orbitals are less accessible for the charge carriers. Consequently, the region A is much less metallic than in case of Tl-2201 and the planar-oxygen mode manifests itself rather as a peak than as a step feature. This is demonstrated in Fig. 5 (d), where the results for $\Omega_A = 1500$ cm$^{-1}$ (dotted line) and $\Omega_A = 600$ cm$^{-1}$ (dashed-dotted line) are shown.

An observation of the infrared anomalies discussed above in more precise future experiments would represent a clear but indirect evidence in favour of the suggested extension of the ILT theory. Direct evidence could be provided by experiments probing the occupation of individual atomic orbitals, as, e.g., the near x-ray absorption fine structure (NEXAFS) measurements (see, e.g., Ref. [71]). We predict that such experiments will reveal a sizeable increase of the number of holes on the apical sites at $T_c$ accompanied by the corresponding decrease of the number of holes in the copper-oxygen planes. Note that this prediction does not concern only Tl-2201 but all the single layer high-$T_c$ cuprates. It may, of course, also be fulfilled for some of the bilayer cuprates. The changes of the hole distribution at $T_c$ should further result in some structural changes, in particular, in changes of the distance between the apical oxygens and the copper-oxygen planes, and in anomalies of some Raman-active phonon modes involving vibrations of the apical oxygens. Such anomalies may already have been observed [2].

VI. SUMMARY

The $c$-axis infrared conductivity of optimally doped Bi-2212 exhibits the same kind of anomalies as that of underdoped Y123. Below $T_c$ the electronic background increases in the frequency region around 550 cm$^{-1}$ and at the same time the oxygen bond-bending mode at 355 cm$^{-1}$ loses a large part of its spectral weight. The anomalies can be explained within a model involving the intra-bilayer Josephson effect and variations of the electric field inside the unit cell. We have compared the Josephson-coupling energies of Y123 and Bi-2212 with different oxygen concentrations obtained from the optical data with the condensation energies obtained from the specific heat data and we have found that there is a remarkable agreement between the values of the two quantities. The Josephson coupling energy is shown to represent a reasonable estimate of the change of the $c$-axis kinetic energy upon entering the superconducting state and it is also shown that the latter quantity cannot be obtained by using the simple “tight-binding” sum rule, since this sum rule has been derived assuming homogeneous distribution of the total electric field within the unit cell. The most plausible interpretation of the agreement between the Josephson coupling energies and the condensation
energies is that the condensation energy of the bilayer compounds can be accounted for by the interlayer tunneling theory. We propose a modification of this theory which may also explain the high values of $T_c$ in Tl-2201 and Hg-1201. The main idea is that the $c$-axis kinetic energy of these compounds decreases at $T_c$ via a delocalization of the Cooper pairs onto the apical-oxygen orbitals. We have investigated, using a toy model, the consequences of this hypothesis for the $c$-axis infrared response and we have demonstrated that it offers a simple explanation of two features observed in the measured $c$-axis conductivity of Tl-2201. In addition, we propose that for Tl-2201 an increase of the electronic background around 1000 cm$^{-1}$ takes place at $T_c$. We further predict a sizeable increase at $T_c$ of the number of holes on the apical sites, related structural changes and related anomalies of some Raman-active phonon modes.

Note added. There are some similarities between our model [18] of the charge dynamics and the models of Refs. [73,74,75,76,77]. In Ref. [73] it is argued that the role of the intra-bilayer plasmon could be played by an antiphase oscillation between the copper-oxygen planes and the ordered blocking layer plaques. After submitting the paper, O. K. Andersen has drawn our attention to a preprint of Pavarini et al. [78], where it is shown that there is a correlation between the maximum value of $T_c$ for a given compound, $T_{c\text{max}}$, and the value of the parameter $r$ which expresses the range of the intra-layer hopping, $t'/t$, obtained from band structure calculations. The higher the value of $r$, the higher the value of $T_{c\text{max}}$. It remains an open question to what extent this correlation is consistent with the extension of the ILT theory proposed in Sec. V. There we have suggested that the condensation energy of the single layer compounds is largely due to a change of the $c$-axis kinetic energy at $T_c$ connected with an increase of the number of holes on the apical sites. The reason for this change is the confinement of the holes to the copper oxygen planes in the normal state and a deconfinement of the Cooper pairs below $T_c$. In case of the single layer systems the condensation energy—and also $T_c$—thus should be the largest (ignoring the material dependence of the orbital energies) for the compounds with the largest value of the hopping matrix element $t_{pa}$ between the relevant orbital of a copper-oxygen plane (predominantly $d_{x^2-y^2}$) and the $2p_z$ orbital of the neighbouring apical oxygen plane. The $2p_z$ orbital of an apical oxygen couples to the copper $4s$ orbital and $t_{pa}$ is thus determined (a) by the matrix element $t_{sc}$ between the $2p_z$ orbital and the Cu $4s$ orbital and (b) by the ratio of the Cu $4s$ to Cu $d_{x^2-y^2}$ character, $R$. The value of $t_{sc}$ decreases [73] with increasing distance $d$ between the copper-oxygen plane and the apical-oxygen plane and it is thus somewhat smaller for Tl-2201 ($d \approx 2.7\AA$) and Hg-1201 ($d \approx 2.8\AA$) as compared to LaSrCuO$_4$ ($d \approx 2.4\AA$). On the other hand, the value of $R$ increases with increasing value of $r$ as $r^2$ [78], which means that it is much larger for Tl-2201 ($r \approx 0.33$) and Hg-1201 ($r \approx 0.33$) than for LaSrCuO$_4$ ($r \approx 0.17$) [78]. For this reason it is possible that $t_{pa}$ increases with increasing $r$, i.e., that the correlation reported in Ref. [78] is consistent with what one would expect on the basis of the extended ILT theory. Obviously, a careful analysis involving the details of the Cu $4s$—apical-oxygen $2p_z$ bonding is required.

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Appendix A: The main ideas of the model of Ref. [18]

Figure. 7 (a) shows a schematic representation of the electronic part of the model of Ref. [18]. The effects related to the ionic degrees of freedom will be discussed subsequently. Thick horizontal lines correspond to the two-dimensional copper-oxygen planes. The distance between the closely spaced copper-oxygen planes forming a bilayer is denoted by $d_{bl}$, the distance between the bilayers is denoted by $d_{int}$. If an electric field $E'$ is applied, the currents $j_{bl}$ (intra-bilayer current) and $j_{int}$ (inter-bilayer current) flow between the planes. Since they are not equal, the planes become charged. The resulting surface charge density which alternates from one plane to the other is denoted by $\kappa$. It modifies the average electric field in the intra-bilayer region, $E_{bl} = E' + (\kappa/\varepsilon_{0}\varepsilon_{\infty})$, whereas it does not change the average electric field in the inter-bilayer region, $E_{int} = E'$. The currents $j_{bl}$ and $j_{int}$ are determined by the fields $E_{bl}$ and $E_{int}$, respectively, $j_{bl} = \sigma_{bl}E_{bl}$ and $j_{int} = \sigma_{int}E_{int}$, so that a selfconsistent set of equations is obtained. Here $\sigma_{bl}$ and $\sigma_{int}$ are the intra- and the inter-bilayer conductivities, respectively. The equations can be readily solved yielding the
located in the intra-bilayer and in the inter-bilayer regions experience then the electric fields acting on these ions is then 
\( E = \frac{\varepsilon_{\text{bl}}(\omega) + \varepsilon_{\text{int}}(\omega)}{\varepsilon(\omega)} \).

Here \( \varepsilon_{\text{bl}}(\omega) = (i/\varepsilon_0 \omega) \sigma_{\text{bl}}(\omega) \) and \( \varepsilon_{\text{int}}(\omega) = (i/\varepsilon_0 \omega) \sigma_{\text{int}}(\omega) \). For a superlattice of intra-bilayer and inter-bilayer Josephson junctions we have \( \varepsilon_{\text{bl}}(\omega) = \varepsilon_{\infty} - (\omega_0^2/\omega^2) \) and \( \varepsilon_{\text{int}}(\omega) = \varepsilon_{\infty} - (\omega_0^2/\omega^2) \). The dielectric function of Eq. (A.1) then exhibits two conventional plasma resonances at \( \omega = \omega_{\text{bl}}/\sqrt{\varepsilon_{\infty}} \) and at \( \omega = \omega_{\text{int}}/\sqrt{\varepsilon_{\infty}} \). In addition it exhibits a pole at

\[
\omega = \omega_p = \sqrt{\frac{d_0 \omega_{\text{int}}^2 + d_{\text{int}} \omega_{\text{bl}}^2}{(\varepsilon_{\text{bl}} + \varepsilon_{\text{int}}) \varepsilon_{\infty}}},
\]

corresponding to a resonant oscillation of the condensate density between the two closely spaced copper-oxygen planes (transverse plasma excitation). The spectral weight of the \( \delta \)-peak at \( \omega = 0 \) is

\[
S_\delta = \frac{(\pi/2)\varepsilon_0 (d_0 + d_{\text{int}}) \omega_{\text{int}}^2 \omega_{\text{bl}}^2}{(d_0 + d_{\text{int}}) \omega_{\text{int}}^2 + d_{\text{int}} \omega_{\text{bl}}^2},
\]

and the spectral weight of the resonance at \( \omega_p \) is

\[
S_{\text{pl}} = \frac{(\pi/2)\varepsilon_0 (d_0 + d_{\text{int}}) (\omega_{\text{int}}^2 - \omega_{\text{bl}}^2)^2}{(d_0 + d_{\text{int}}) \omega_{\text{int}}^2 + d_{\text{int}} \omega_{\text{bl}}^2}.
\]

Equations (A.1) and (A.2) have been previously derived in another way by Van der Marel and Tsutsumi (see also Ref. [13]). As far as only the electronic degrees of freedom are concerned, the two approaches yield completely equivalent results. However, the present approach can be more easily extended to incorporate the phonons.

Assume for a moment that the displacements of the ions do not modify the electric fields \( E_{\text{bl}} \) and \( E_{\text{int}} \). The ions located in the intra-bilayer and in the inter-bilayer regions experience then the electric fields \( E_{\text{bl}} \) and \( E_{\text{int}} \), respectively. The ions located in the copper oxygen planes experience a field \( E_{\text{loc}} \) which is equal to the average of the two fields, \( E_{\text{loc}} = (E_{\text{bl}} + E_{\text{int}})/2 \). This can be easily explained using Fig. 7 (a). Let us consider, e.g., the ions in the middle copper-oxygen plane. They experience the applied field \( E' \) and the electric field generated by the charge density \( -\kappa \) of the upper copper-oxygen plane (the contributions of the other planes cancel each other). The total electric field acting on these ions is then \( E_{\text{loc}} = E' + (\kappa/2\varepsilon_0 \varepsilon_{\infty}) = (E_{\text{bl}} + E_{\text{int}})/2 \). Even this simplified picture of the electric fields acting on the ions allows us to explain the spectral weight anomalies. The spectacular reduction of the spectral weight of the 320 cm\(^{-1} \) phonon mode in underdoped Y123 shown in Fig. 1, e.g., is due to the fact that in the frequency region around the phonon the two fields \( E_{\text{bl}} \) and \( E_{\text{int}} \) have opposite signs (see Ref. [18] for a discussion).

In reality, the displacements of the ions modify the electric fields \( E_{\text{bl}} \) and \( E_{\text{int}} \), so that the model equations become slightly more complicated. As an example, we show in Fig. 7 (b), how the displacement of the planar oxygens of Y123 (O(2) and O(3)) from their equilibrium position influences the electric field \( E_{\text{bl}} \). We refer the reader to Refs. [18,30] for further details of the model.

**Appendix B: Approximate “tight-binding” sum rule for bilayer compounds**

In this appendix we put the conclusions of section IV B on a more rigorous basis and we present a way of estimating the contribution of the countereffects [5]. We shall discuss a microscopic counterpart of the model outlined in Appendix A. For this reason, it may be helpful for the reader to follow Fig. 7. Let us consider a model defined by the following hamiltonian

\[
H = H_{\text{in-plane}} + H_{\text{bl}} + H_{\text{int}},
\]

where \( H_{\text{in-plane}} \) contains the intra-planar single particle terms and interaction terms, \( H_{\text{bl}} \) and \( H_{\text{int}} \) contain the hopping terms between the closely-spaced copper-oxygen planes and between the widely spaced ones, respectively,

\[
H_{\text{bl}} = -t_{\perp \text{bl}} \sum_{l=1,3,5,...;i,s} c_{i,l+1,s}^\dagger c_{i,l,s} + H.c.,
\]

\[
H_{\text{int}} = -t_{\perp \text{int}} \sum_{l=2,4,6,...;i,s} c_{i,l+1,s}^\dagger c_{i,l,s} + H.c.,
\]

for further details of the model.
In the presence of an electric field along the c-axis, the hamiltonian reads:

\[ H_A = H - Na^2 \sum_{l=1,3,5, \ldots} \left( d_{bl} j^p(l) A_{bl} + e^2 d_{bl}^2 2a^2 h^2 k(l) A_{bl}^2 \right) - Na^2 \sum_{l=2,4,6, \ldots} \left( d_{int} j^p(l) A_{int} + e^2 d_{int}^2 2a^2 h^2 k(l) A_{int}^2 \right) \]  

(B.A)

Here

\[ j^p(l) = \frac{i e t_{\perp}}{Na^2 h} \sum_{i,s} \left( c_{i+l+1,s}^+ c_{i+l,s} - c_{i+l,s}^+ c_{i+l+1,s} \right) \]  

(B.5)

where \( \alpha = bl \) for \( l \in \{1, 3, 5, \ldots \} \) and \( \alpha = int \) for \( l \in \{2, 4, 6, \ldots \} \), are the c-axis paramagnetic current densities. Further,

\[ k(l) = \frac{t_{\perp} \alpha}{N} \sum_{i,s} \left( c_{i+l+1,s}^+ c_{i,l,s} + c_{i,l,s}^+ c_{i+l+1,s} \right) \]  

(B.6)

are the c-axis kinetic energies (per unit cell), \( N \) is the number of lattice sites in one copper-oxygen plane, \( A_{bl} \) is the vector potential in the intra-bilayer region \( (A_{bl} = i \omega E_{bl}) \), \( A_{int} \) is the vector potential in the inter-bilayer region \( (A_{int} = i \omega E_{int}) \). When studying the optical response of the system, selfconsistent values of these two vector potential have to be used (RPA approximation). Using similar manipulations as in Ref. [50], we obtain the following relation between the averaged total current densities, \( j_\alpha \), and the two electric fields, \( E_\alpha \),

\[ j_\alpha = \sigma_{\alpha,\beta} E_\beta . \]  

(B.7)

Here

\[ j_\alpha = \langle j^p(l) + \frac{e^2 d_{\alpha}^2 A_\alpha}{\hbar^2 a^2 d_\alpha} k(l) \rangle , \]  

(B.8)

where \( l = 1 \) for \( \alpha = bl \) and \( l = 2 \) for \( \alpha = int \). Note that all intra-bilayer regions are identical and all inter-bilayer regions are identical, the long-wavelength limit is assumed. Finally the four conductivities \( \sigma_{\alpha,\beta} \) \((\alpha, \beta \in \{bl, int\})\) are given as follows:

\[ \sigma_{\alpha,\beta}(\omega) = \frac{\langle k(l) \rangle e^2 d_{\alpha} \delta_{\alpha,\beta} / (\hbar^2 a^2) + Na^2 d_{\alpha} T_{\alpha,\beta}(\omega)}{i(\omega + i\delta)} . \]  

(B.9)

Here

\[ T_{\alpha,\beta}(\omega) = i \frac{1}{\hbar} \sum_{l} \int_{-\infty}^{\infty} dt \Theta(t) \Theta(t') \langle [j^p(l, t), j^p(l', t')] \rangle e^{i\omega(t-t')} , \]  

(B.10)

where \( l = 1 \) for \( \alpha = bl \) and \( l = 2 \) for \( \alpha = int \); the sum runs over the odd values of \( l' \) for \( \beta = bl \) and over the even values of \( l' \) for \( \beta = int \). We are not going to discuss the general case here. Instead we concentrate on the case where \( t_{\perp int} < t_{\perp bl} \), i.e., the case of negligible inter-bilayer kinetic energy \( \langle k(2) \rangle \). In this case we can neglect \( \sigma_{bl,int}, \sigma_{int,bl} \) and \( \sigma_{int,int} \) and we have \( j_{bl} \approx \sigma_{bl,bl} E_{bl}, j_{int} \approx 0 \). The current-current correlation functions of Eq. (B.10) have the analytic properties required for the validity of the sum rules and we obtain

\[ \int_{0}^{\infty} \sigma_{bl}(\omega) d\omega = -\frac{\pi e^2 d_{bl}}{2\hbar^2 a^2} \langle k(1) \rangle , \]  

(B.11)

where \( \sigma_{bl}(\omega) \equiv \sigma_{bl,bl}(\omega) \). The sum rule (B.11) yields a formula for the change of the kinetic energy upon entering the superconducting state analogous to Eq. (8) or Eq. (10) \( (\sigma(\omega) \) is substituted by \( \sigma_{bl}(\omega) \) and \( d \) is substituted by \( d_{bl} \)). For vanishingly small values of the regular part of \( \sigma_{bl} \), we obtain \( \Delta \langle k(1) \rangle [\text{meV}] = -4C \left( \omega_{bl} [\text{cm}^{-1}] \right)^2 / d_{bl} [\text{Å}] \). This is nothing else than Eq. (3) except for the factor of 4 [56]. In a general case, Eq. (11) yields a recipe for how to treat the countereffects.
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[42] S. Tajima, G. D. Gu, S. Miyamoto, A. Odagawa, N. Koshizuka, Phys. Rev. B 48, 16164 (1993). The broadening of the 600 cm$^{-1}$ phonon mode apparent in Fig. 3 of this paper may correspond to the additional spectral weight identified in our investigations.

[43] H. Shibata, A. Matsuda, Phys. Rev. B 59, 11 672 (1999), and references therein.

[44] For Bi-2212,

\[ \alpha = -\frac{[n_{Ca}e_Ca + (n_{Ca}e_{Ca}/2)](d_A + d_B)/|n_{O(1)}e_{O(1)}d_A|}{\beta} = \frac{n_{Ba}e_{Ba} + n_{O(3)}e_{O(3)} + n_{Ba}e_{Ba} + n_{O(2)}e_{O(2)} + (n_{Ca}e_{Ca}/2)](d_A + d_B)/|n_{O(1)}e_{O(1)}d_A|}, \]

\[ \gamma = (d_A + d_B)/|d_A|, \]

where, e.g., \( n_{Ca} \) is the number of Ca ions per unit cell, O(1) denotes the planar oxygen, O(3) the BiO-layer oxygen, and O(2) the apical oxygen. For Tl-2201,

\[ \alpha = -\frac{n_{Ba}e_{Ba} + n_{O(3)}e_{O(3)} + n_{Ba}e_{Ba} + n_{O(2)}e_{O(2)} + (n_{Ca}e_{Ca}/2)](d_A + d_B)/|n_{O(1)}e_{O(1)}d_A| = 1.1, \]

\[ \beta = -\frac{n_{TI}e_{TI} + n_{O(1)}e_{O(1)}](d_A + d_B)/|n_{O(2)}e_{O(2)}d_B| = 0.9, \]

\[ \gamma = (d_A + d_B)/d_A = 1.8, \]

where O(1) denotes the TIO-layer oxygen, O(2) the apical oxygen, and O(3) the planar oxygen. These values correspond to the case where the region A is considered as “intra-bilayer”. In the other case (A is considered as “inter-bilayer”) the value of \( \alpha (\beta) \) corresponds to the above value of \( \beta (\alpha) \) and \( \gamma = (d_A + d_B)/d_A = 2.2. \)

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[46] The formula is equivalent to formula (5) of Ref. 3 or formula (1) of Ref. 4. It differs by a factor of 4 from formula (4) of Ref. 4.

[47] The values of \( \omega_{pi} \) can be simply guessed from the data by inserting the frequency of the transverse plasma excitation into Eq. (A.2) and using an estimate of \( \varepsilon_{\infty} \). However, the plasma frequency is screened not only by the interband processes contributing to \( \varepsilon_{\infty} \) but also by midinfrared processes. The description of the contribution of these processes contains a certain degree of arbitrariness. In particular, when fitting the infrared data, one has to make some assumptions concerning the frequency dependence of this contribution and concerning its distribution between the intra- and inter-bilayer regions. Note that the assumptions made in Ref. 3 are different from those made in Ref. 4.

[48] In Ref. 3 we have suggested that the onset of the anomalies above \( T_c \) can be explained by assuming that the intraband plasmon starts to develop already below \( T^* > T_c \). There is no global phase coherence above \( T^*_c \) and consequently, strictly speaking, no Josephson effects. It can be speculated, however, that above \( T_c \) there is either a certain degree of phase coherence between the closely spaced copper oxygen planes or at least some kind of pairing developed making a (semi)coherent intra-bilayer hopping possible.

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[53] This scale corresponds to the frequency range where the single particle tunneling is completely blocked.

[54] In reality the regions of intraband and interband absorption overlap. Considering this would make the discussion more complicated, while it would not change our main conclusions.

[55] The contribution to the c-axis kinetic energy due to the single particle tunneling may increase below \( T_c \); that is what we mean by “countereffects”.

[56] For a given plasma frequency of the condensate (or, alternatively, for a given penetration depth) the sum rule-based-approach of Refs. 24 (Eq. (8)) yields a change of the c-axis kinetic energy by a factor of 4 larger than Eq. (3) if the changes of the regular part of \( \sigma_1(\omega) \) between the normal and the superconducting state are negligible.

For undoped \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) this discrepancy is partially reduced because of the changes in the background.

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[60] This is particularly clear in case of undoped Y123 with \( T_c = 53 \text{K} \) (see Fig. 1 (a)). Both the additional peak and the anomaly of the 320 cm$^{-1}$ phonon mode can be fitted using only three essential parameters: \( \omega_{pi} \), \( S_P \) (oscillator strength of the phonon) and \( \omega_P \) (frequency of the phonon).

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It is surprising that the Tl-layer-oxygen mode exhibits the anomalies rather than the apical-oxygen mode (the apical-oxygen plane forms a boundary between the regions A and B in the same way as the plane of the planar oxygens in Y123 forms a boundary between the intra-bilayer and the inter-bilayer regions). The reason is that the geometry is different. In Y123, $d_{\text{int}} > d_B$, here $d_A \approx d_B$.

The values of the model conductivity in the frequency region around 800 cm$^{-1}$ are by a factor of $\sim 2$ higher than the value of $\sim 10 \Omega^{-1}$ cm$^{-1}$ reported in Ref. [23]. This is not a serious discrepancy considering the oversimplified ansatz for $\sigma_A$ and $\sigma_B$.

The value of the condensation energy of Tl-2201 can be estimated using the specific heat data presented in J. M. Wade, W. J. Loram, K. A. Misra, J. R. Cooper, J. L. Tallon, J. Superconductivity 7, 261 (1994). The value of $\delta \gamma(T_c)$ is by a factor of $\sim 3$ smaller than that for optimally doped Y123 suggesting $U_0 \approx 0.1$ meV.

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### TABLE I. Values of the fitting parameters and other numerical factors used in fitting the measured $\varepsilon$-axis infrared conductivity of optimally doped Bi-2212. The plasma frequency and the broadening parameter of the broad Drude peak are denoted by $\Omega_{bl}$ and $\gamma_{bl}$, respectively. The meaning of the other symbols is the same as in Ref. [18]. The temperatures are given in K, the frequencies and the broadening parameters in cm$^{-1}$. The values of the numerical factors $\alpha$, $\beta$, $\gamma$ [18], $\alpha = 2.28$, $\beta = 0.64$, $\gamma = 1.28$ have been obtained in the same way as in Ref. [18] (see also Ref. [44]) using the following values of the effective ionic charges: $e_{Sr}^* = 3$, $e_{Cu}^* = 2$, $e_{Ca}^* = 2$, $e_{O}^* = -2$. The distances between the planes of a bilayer and between the neighbouring bilayers are $d_{bl} = 3.37 \text{ Å}$ and $d_{int} = 12.03 \text{ Å}$, respectively.

| $T$ (K) | $\varepsilon_\infty$ | $\omega_{bl}$ | $\Omega_{bl}$ | $\gamma_{bl}$ | $S_{bl}$ | $\omega_0$ | $\gamma_0$ | $S_P$ | $S_1$ | $S_2$ | $\omega_P$ | $\omega_1$ | $\omega_2$ | $\gamma_P$ | $\gamma_1$ | $\gamma_2$ |
|-------|-------------------|---------------|---------------|---------------|---------|---------|---------|------|------|------|---------|---------|---------|---------|-------|------|
| 300   | 3.8               | 0             | 3000          | 1220          | 0       | 0       | 1.05    | 0.24 | 0.50 | 455  | 332     | 639     | 10      | 36      | 31    |
| 100   | 3.8               | 0             | 3000          | 1300          | 0       | 0       | 1.05    | 0.24 | 0.50 | 455  | 332     | 639     | 12      | 35      | 33    |
| 10    | 3.8               | 1180          | 2520          | 1300          | 2.04    | 1000    | 900     | 1.05  | 0.24 | 0.50 | 455     | 332     | 636     | 12      | 34    | 36    |

### TABLE II. Values of the intra-bilayer plasma frequency $\omega_{bl}$, the Josephson coupling energy $E_J$ and the condensation energy $U_0$ for several bilayer compounds. The values of $\omega_{bl}$ are taken from Ref. [18] (underdoped Y123), Ref. [19] (optimally doped Y123), Ref. [21] (underdoped Bi-2212); the value for optimum doped Bi-2212 has been obtained as described in the text. The values of the condensation energies are taken from Ref. [22] (Y123) and Ref. [23] (Bi-2212).

| compound | $T_c$ (K) | $\omega_{bl}$ (cm$^{-1}$) | $E_J$ (meV) | $U_0$ (meV) | $E_J/U_0$ |
|----------|-----------|--------------------------|-------------|-------------|-----------|
| YBa$_2$Cu$_3$O$_{6.45}$ | 25         | 950                      | 0.08        | 0.01        | 8.0       |
| YBa$_2$Cu$_3$O$_{6.53}$ | 53         | 1200                     | 0.13        | 0.05        | 2.6       |
| YBa$_2$Cu$_3$O$_{6.75}$ | 80         | 1780                     | 0.30        | 0.16        | 1.8       |
| YBa$_2$Cu$_3$O$_{6.93}$ | 91         | 3480                     | 1.14        | 0.36        | 3.2       |
| Bi$_2$Sr$_2$CaCu$_2$O$_8$ | 60         | 620                      | 0.035       | 0.02        | 1.5       |
| Bi$_2$Sr$_2$CaCu$_2$O$_8$ | 80         | 970                      | 0.085       | 0.06        | 1.4       |
| Bi$_2$Sr$_2$CaCu$_2$O$_8$ | 91         | 1180                     | 0.13        | 0.13        | 1.0       |
TABLE III. Values of the parameters and other numerical factors used in modelling the c-axis infrared conductivity of Tl-2201. The plasma frequency and the broadening parameter of $\sigma_A$ and $\sigma_B$ are denoted by $\Omega_A$ and $\gamma_A$ and $\Omega_B$ and $\gamma_B$, respectively. The parameters of the phonons are denoted by $S_1$, $\omega_1$, $\gamma_1$, $S_2$, $\omega_2$, $\gamma_2$, $S_3$, $\omega_3$, $\gamma_3$. The frequencies and the broadening parameters are given in cm$^{-1}$. The values of the numerical factors $\alpha$, $\beta$, $\gamma$ have been obtained in the same way as in Ref. [13] (see also Ref. [14]) using the following values of the effective ionic charges: $\epsilon_{A_{1}} = 3$, $\epsilon_{Ba} = 2$, $\epsilon_{Cu} = 2$, $\epsilon_{O} = -2$. The distances between the apical-oxygen planes are $d_{A} = 5.3$ Å and $d_{B} = 6.3$ Å, respectively.

| $\varepsilon_{\infty}$ | $\Omega_{A_{1}}$ | $\Omega_{A_{2}}$ | $\gamma_{A}$ | $\Omega_{B}$ | $\gamma_{B}$ | $S_1$ | $S_2$ | $S_3$ | $\omega_1$ | $\omega_2$ | $\omega_3$ | $\gamma_1$ | $\gamma_2$ | $\gamma_3$ |
|------------------------|------------------|------------------|-------------|-------------|-------------|------|------|------|----------|----------|----------|------|------|------|
| 5                      | 2500             | 5000             | 2000        | 600         | 2000        | 0.6  | 0.5  | 0.5  | 300      | 440      | 640      | 25   | 20   | 25   |

FIG. 1. (a) Experimental spectra of the c-axis conductivity, $\sigma = \sigma_1 + i\sigma_2$, of Y123 with $T_c = 53$ K. Experimental data (thick solid lines) and fits (thin solid lines) for (b) $T = 300$ K and (c) $T = 4$ K. The symbols (A), (B), and (C) indicate the most pronounced anomalies as discussed in the text. A slightly modified version of Fig. 2 from Ref. [30].

FIG. 2. Experimental spectra of the c-axis conductivity of almost optimally doped Bi-2212 with $T_c = 91$ K. (a) Data for $T = 300$ K and $T = 100$ K. (b) Data for $T = 100$ K and $T = 10$ K. The insets of (a) and (b) display the differences $\sigma_1(T = 100$ K) $- \sigma_1(T = 300$ K) and $\sigma_1(T = 10$ K) $- \sigma_1(T = 100$ K), respectively. Note that the former and the latter difference corresponds to a temperature change of 200 K and 90 K, respectively. The symbols (A) and (B) in (b) indicate the most pronounced anomalies as discussed in the text.

FIG. 3. (a) Experimental spectra of the c-axis conductivity of optimally doped Bi-2212—region of the spectra near the anomalies on an enlarged scale. (b) Fits of the spectra obtained by using the model of Ref. [18] as described in the text. (c) Spectra of $\sigma_1(T = 100$ K) $- \sigma_1(T = 10$ K). (d) Spectra of the quantity $N_0 - N_1$ defined in the text.

FIG. 4. Schematic representation of the spectral-weight changes upon entering the superconducting state. The increase of spectral weight at low frequencies below $T_c$ is compensated by a decrease around some frequency $\omega_c$. This frequency may be either (a) higher or (b) lower than the interband cutoff $\omega_c$. The horizontal lines below the frequency axis of (b) indicate the three frequency scales corresponding to the three effective c-axis kinetic energies discussed in the text.

FIG. 5. Results of model computations of the c-axis infrared conductivity of Tl-2201. The results shown in (a) and (b) have been obtained using the assignment $A_{1}$ (the Tl-layer oxygens vibrate at the highest frequency, the apical oxygens at $\sim 400$ cm$^{-1}$), those shown in (c) and (d) correspond to the reverse assignment $A_{2}$. The results shown in (a) and (c) have been obtained considering the region A as “intra-bilayer”, those shown in (b) and (d) have been obtained considering the region A as “inter-bilayer”. The planar oxygen mode manifests itself as a step around 240 cm$^{-1}$ in (b) and (d). The dashed and solid lines correspond to the two values of the plasma frequency of region A, $\Omega_{A_{1}}$ and $\Omega_{A_{2}}$, $\Omega_{A_{1}} < \Omega_{A_{2}}$, respectively, as discussed in the text. The dotted and dashed-dotted lines in (d) represent results for $\Omega_{A} = 1500$ cm$^{-1}$ and $\Omega_{A} = 600$ cm$^{-1}$, respectively. The inset of (b) displays the step in the experimental data of Ref. [29]. The inset of (c) displays the experimental spectra of $(N_0 - N_1)/\rho_s$ taken from Ref. [29]. The arrow demonstrates the increase of the spectral weight of the phonon peak at 390 cm$^{-1}$ in the superconducting state, as discussed in the text.

FIG. 6. Crystal structure of Tl$_2$Ba$_2$CuO$_6$.

FIG. 7. (a) Schematic representation of the model. (b) The average electric field $E_{bl}$ between the charged planes corresponding to the planar oxygens (thick horizontal lines) displaced from their equilibrium positions (thin horizontal lines) possesses a contribution $\Delta E_{bl}$ due to the charged Y-plane (dashed line), $\Delta E_{bl} = [(d_{bl}/2) + a]E_Y + ((d_{bl}/2) - a)(-E_Y)]/d_{bl}$. Here $E_Y$ is the field due to the Y-plane in the upper region of the figure. A slightly modified version of Fig. 2 from Ref. [30].
conductivity ($\Omega^{-1}\text{cm}^{-1}$)

$\sigma_1(T=300 \text{ K})$

$\sigma_1(T=4 \text{ K})$

$\sigma_2(T=300 \text{ K})$

$\sigma_2(T=4 \text{ K})$

$\nu$ (cm$^{-1}$)

(a) $\sigma_1(T=300 \text{ K})$

$\sigma_1(T=4 \text{ K})$

$\sigma_2(T=300 \text{ K})$

$\sigma_2(T=4 \text{ K})$

(b) $T=300 \text{ K}$

(c) $T=4 \text{ K}$
Bi2212

(a) Bi2212

σ [Ω⁻¹ cm⁻¹]

ν [cm⁻¹]

0 100 200 400 500 600

(b) Bi2212

σ [Ω⁻¹ cm⁻¹]

ν [cm⁻¹]

0 100 200 300 400 500 600

∆σ [Ω⁻¹ cm⁻¹]

ν [cm⁻¹]
(a) \( T = T_c \)

\( T \ll T_c \)

\( \delta \)-peak

(\( \sigma \) vs. \( \omega \))

(b) \( T = T_c \)

\( T \ll T_c \)

\( \delta \)-peak

(\( \sigma \) vs. \( \omega \))
$\text{Tl}_2\text{Ba}_2\text{CuO}_6$

CuO$_2$ plane
Apical oxygen plane

Region A
Region B

- **Tl**
- **Ba**
- **Cu**
- **O**
