

## **c**-axis response of $\text{YBa}_2\text{Cu}_4\text{O}_8$ : A pseudogap and possibility of Josephson coupling of $\text{CuO}_2$ planes

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We report on the temperature dependence of the far-infrared conductivity of  $\text{YBa}_2\text{Cu}_4\text{O}_8$  single crystals for  $\mathbf{E}\parallel c$ . With decreasing temperature, the conductivity shows a transition from a Drude-like behavior to a pseudogap at  $180\text{ cm}^{-1}$  which grows deeper below 180 K without any abrupt changes at the superconducting transition at  $T_c=80\text{ K}$ . In the superconducting state the formation of the superfluid condensate can be seen. We show that in a variety of high- $T_c$  superconductors the value of the London penetration depth for  $\mathbf{E}\parallel c$  correlates with the conductivity along the  $c$  direction. With respect to this correlation we discuss the possibility of Josephson coupling between the  $\text{CuO}_2$  layers.

The electromagnetic response of high- $T_c$  superconductors is strongly anisotropic offering a striking contrast between the nearly insulating  $c$  direction and the well conducting  $ab$  plane.<sup>1-8</sup> Recently, it has been shown that the semi-insulating behavior of the  $c$ -axis properties in the underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{6.70}$  is the result of the formation of a pseudogap in the real part of the conductivity  $\sigma_1(\omega)$ .<sup>9</sup> The closely related material,  $\text{YBa}_2\text{Cu}_4\text{O}_8$  is naturally underdoped as well as untwinned and has been reported to show evidence of a gap in NMR measurements<sup>10,11</sup> as well as in the  $a$ -axis resistivity.<sup>12</sup> In this paper we report on the temperature dependence of the  $c$ -axis far infrared (FIR) conductivity which shows the opening of a pseudogap at temperatures well above the superconducting transition at 80 K. We discuss the formation of the superfluid at  $T < T_c$ , as seen in the  $c$ -axis response, in terms of the Josephson coupling between  $\text{CuO}_2$  layers. Within this model we are able to describe the observed correlation between the  $c$ -axis London penetration depth  $\lambda_c$  and the  $c$ -axis conductivity in a variety of high- $T_c$  materials.

The details of sample preparation and characterization were reported elsewhere.<sup>13</sup> The typical size of the crystals was  $500 \times 500 \times 60\ \mu\text{m}^3$ , and in order to obtain an area of sufficient size for FIR spectroscopy, four samples were mounted together. The reflectance was measured between 70 and  $5000\text{ cm}^{-1}$ . To account for the finite sample size and surface irregularities the sample was coated *in situ* with a gold layer and the spectrum of the gold-coated sample was then used as a reference.<sup>14</sup> The uncertainty of the absolute reflectivity was estimated to be about 1%.

In Fig. 1 we show the reflectivity spectra in the FIR region at several different temperatures. The response is that of a poor metal with additional contributions from IR active phonon modes. In the normal state, the reflectivity *decreases* as the temperature is lowered, contrary to what is expected

for a metal. At temperatures below  $T_c$  a plasma edge due to the formation of superfluid is clearly seen in the reflectivity.

The optical conductivity, determined through Kramers-Kronig analysis of the reflectance, is plotted in Fig. 2. The Hagen-Rubens formula and the two-fluid model have been used as a low-frequency extrapolation in the normal and superconducting states, respectively. Five IR active phonon modes at frequencies 132, 193, 303, 493, and  $593\text{ cm}^{-1}$  dominate the conductivity. Of these, the three high-frequency modes originate from oxygen vibrations<sup>15,16</sup> and have an asymmetric line shape which is strongly temperature dependent.

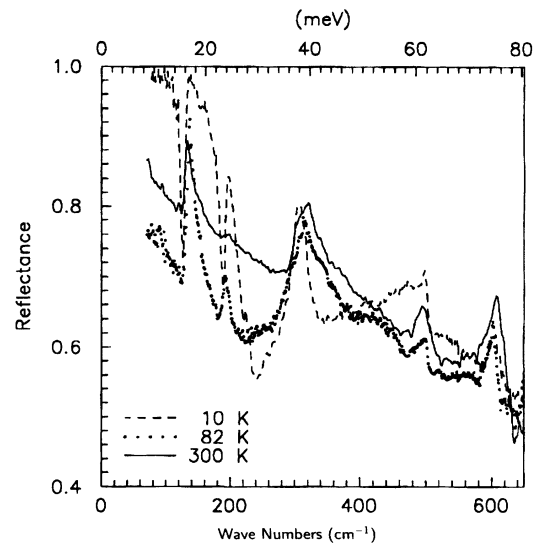


FIG. 1. Reflectivity spectra of  $\text{YBa}_2\text{Cu}_4\text{O}_8$  for  $\mathbf{E}\parallel c$  at different temperatures. Below  $T_c=80\text{ K}$  the reflectance shows a plasma edge at low frequencies.

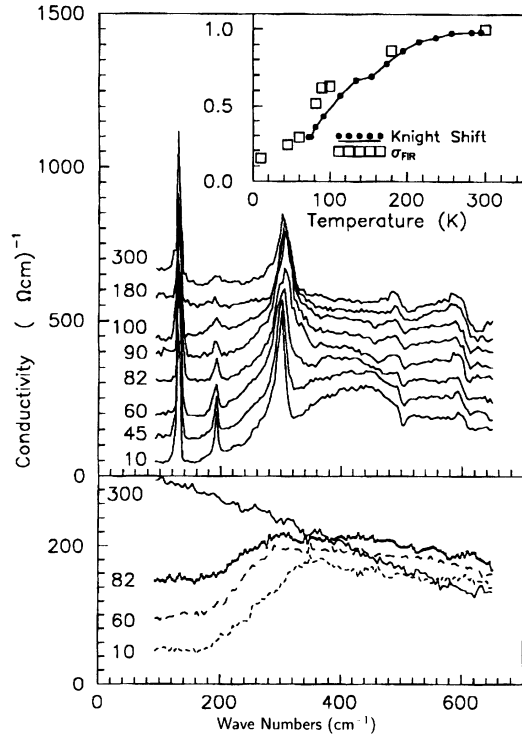


FIG. 2. Top panel:  $c$ -axis conductivity of  $\text{YBa}_2\text{Cu}_4\text{O}_8$  at different temperatures. Starting with 45 K spectrum all successive spectra are shifted up by  $50 (\Omega\text{cm})^{-1}$ . The inset: conductivity in gap region ( $\omega < 180 \text{ cm}^{-1}$ ) and of Knight shift (Refs. 10 and 11) normalized to room temperature. Bottom panel: electronic part of the  $c$ -axis conductivity with phonons being subtracted by fits to oscillators.

dent, indicating that there is an interaction with the electronic background.<sup>17</sup> The oscillator strength of these phonons decreases as the temperature is lowered and is transferred to a new broad mode at  $420 \text{ cm}^{-1}$  which appears at  $T < 100 \text{ K}$ . Similar effects have been seen in underdoped YBCO.<sup>9</sup> The total spectral weight of the phonons remains unchanged as required by the oscillator strength sum rule. Since the phonons can be considered to be an independent channel of conductivity we have subtracted their contribution to the  $\sigma_1(\omega)$  by fitting their line shapes with oscillators. We have not observed the anomalously strong softening of high-frequency modes which has been found in ceramic samples.<sup>18</sup>

The electronic conductivity at room temperature follows the Drude model with a scattering rate of about  $600 \text{ cm}^{-1}$ . As the temperature is lowered the conductivity first remains Drude-like with an increasing scattering rate, but at  $T < 180 \text{ K}$  a pseudogap can be seen to open in the conductivity below  $180 \text{ cm}^{-1}$ . We assume that the spectral weight from the low-frequency part of the conductivity ( $\omega < 200 \text{ cm}^{-1}$ ) is transferred to higher energies above the upper limit of the frequency of our measurements. At low temperatures, when the pseudogap is well developed,  $\sigma_1(\omega)$  is constant from our lowest measured frequencies up to a clear onset at  $\omega = 180 \text{ cm}^{-1}$ . The frequency of the onset is independent of temperature. The temperature dependence of the magnitude of the

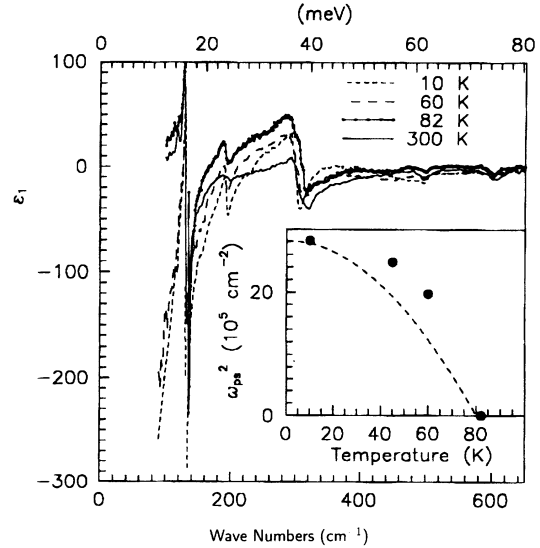


FIG. 3. Frequency dependence of the real part of the dielectric function. In the superconducting state  $\epsilon_1$  is negative and shows a steep slope due to the response of the superfluid.

conductivity in the pseudogap region is plotted in the inset of Fig. 2 and shows a smooth increase of  $\sigma_1$  without any abrupt change at the superconducting  $T_c$ . The observed behavior cannot be attributed to carrier activation across the pseudogap since the dependence flattens at high temperatures. However, this dependence is consistent with the measurements of the Knight shift.<sup>10</sup> Similar properties have been found in the  $c$ -axis response of the oxygen deficient  $\text{YBa}_2\text{Cu}_3\text{O}_{6.70}$  single crystals.<sup>9</sup> There is, however, an important difference between these two systems. In  $\text{YBa}_2\text{Cu}_3\text{O}_{6.70}$  the transition to the pseudogap state occurs from an incoherent conductivity that is featureless, whereas  $\text{YBa}_2\text{Cu}_4\text{O}_8$  shows a transition from a more Drude-like coherent behavior to the pseudogap state. Thus there does not appear to be a direct correlation between the degree of coupling between the planes and the appearance of the pseudogap.

The pseudogap in the far-infrared conductivity of  $\text{YBa}_2\text{Cu}_4\text{O}_8$  develops in the same temperature range where the spin lattice relaxation time,<sup>11</sup> the  $a$ -axis resistivity and the Hall coefficient show an anomalous behavior that has been attributed to the opening of a spin gap.<sup>12</sup>

Though the frequency dependence of the normal state conductivity is characterized by a well-defined pseudogap, in the superconducting state the  $c$ -axis response shows evidence of the formation of superfluid as asserted by the frequency dependence of the real part of the dielectric function  $\epsilon_1$ . In the normal state  $\epsilon_1$  is positive and weakly frequency dependent at low  $\omega$ , suggestive of nonmetallic, or marginally metallic, behavior (Fig. 3). At  $T < T_c$  carrier condensation is indicated by the negative  $\epsilon_1$  with a steep slope. Alternatively, carrier condensation in a superfluid is revealed by a sum rule analysis of the conductivity when part of the spectral weight is missing from the low-frequency region in  $\sigma_1(\omega)$ . The same absolute value of London penetration depth  $\lambda_c$  about  $9000 \text{ \AA}$  at 10 K is found through the sum rule analysis and from  $\epsilon_1$ . Thus it appears that while above  $T_c$  the missing

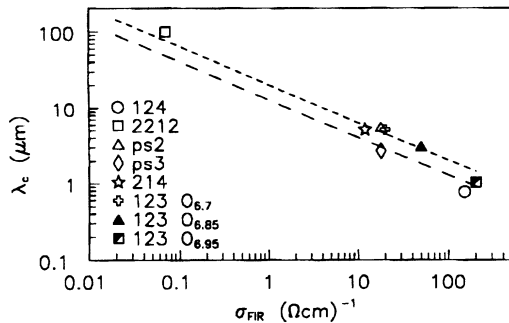


FIG. 4. Correlation between the values of the penetration depth  $\lambda_c$  and the  $c$ -axis conductivity for different high- $T_c$  superconductors (dashed dots): 124—this work, 123—Refs. 9 and 21, 214 ( $\text{La}_{1.84}\text{Sr}_{0.16}\text{CuO}_4$ )—Ref. 6, PS 2 and PS 3  $\text{Pb}_2\text{Sr}_2\text{RCu}_3\text{O}_8$ —Ref. 25. Dashed line—calculation for the case of Josephson coupling between  $\text{CuO}_2$  planes [Eq. (1)] assuming  $2\Delta = 128 \text{ cm}^{-1}$  (short dashes) and  $2\Delta = 320 \text{ cm}^{-1}$  (long dashes).

spectral weight associated with the developing pseudogap goes to high frequencies, below  $T_c$  it goes to the condensate. Similar effects were seen in underdoped YBCO.<sup>9</sup> The temperature dependence of the superfluid plasma frequency  $\omega_{ps} = 1/\lambda_c$  is plotted in the inset of Fig. 3.

In Fig. 4 we show the correlation between the  $c$ -axis penetration depth and the values of  $\sigma_1(\omega \rightarrow 0)$  at  $T = T_c$  in a variety of high- $T_c$  superconductors.<sup>21</sup> In the superconducting state the  $c$ -axis reflectance of all of these materials exhibits a plasma edge at FIR frequencies. In most of these materials the  $c$ -axis conductivity in the normal state shows a nonmetallic response similar to that of  $\text{YBa}_2\text{Cu}_4\text{O}_8$  demonstrated in Figs. 1–3. With the exception of  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  the values of the penetration depth  $\lambda_c$  have been determined from the real part of the dielectric function.<sup>9,22,23,6</sup> In the case of Bi-based material the penetration depth has been determined from the ac susceptibility whereas the  $c$ -axis conductivity has been measured by dc probe.<sup>24</sup> Figure 4 shows that with increasing  $c$ -axis conductivity the penetration depth  $\lambda_c$  decreases systematically revealing a universal behavior of high- $T_c$  superconductors.

The correlation between the penetration depth and the normal state conductivity  $\sigma_n$  can be interpreted in two ways. First, for a bulk superconductor in the dirty limit at low frequencies:

$$\hbar c^2/\lambda^2 = 4\pi\sigma_2(\omega) = 4\pi^2\sigma_n\Delta \tanh(\Delta/2kT), \quad (1)$$

where  $\Delta$  is the energy gap.<sup>25</sup> Equation (1) leads to the proportionality between  $\lambda_c^2$  and the conductivity in the normal state, shown in Fig. 4 by dashed lines. Both the slope and the absolute values of  $\lambda_c$  are reproduced well by Eq. (1) but the conventional dirty limit approach may not be appropriate for the  $c$ -axis response of materials shown in Fig. 4. The conductivity is below Mott's minimum metallic limit and the normal state transport seems to be dominated by the pseudogap in many of the materials shown in Fig. 4.

A second approach is to treat the  $c$ -axis transport as charge transport between superconducting layers coupled by the Josephson effect. It has been suggested by Lawrence and Doniach and by Bulaevski that properties of layered super-

conductors are qualitatively similar to those of a series of Josephson junctions.<sup>26,27</sup> We show below that some of the specific predictions of their model are consistent with the behavior of the penetration depth in high- $T_c$  materials. The in-plane response of all known high- $T_c$  superconductors is metallic suggesting that electronic transport is nearly confined to the quasi-two-dimensional (2D) layers inside the bulk of a crystal (presumably to the  $\text{CuO}_2$  planes). Then 3D superconductivity may be due to the Josephson coupling of the planes. This possibility has been discussed previously in the context of  $c$ -axis optical studies of high- $T_c$  materials.<sup>4,19</sup> A bulk interplane Josephson effect has been observed in the  $c$ -axis transport of the single crystals of  $\text{Bi}_2\text{Ca}_2\text{SrCu}_2\text{O}_8$ .<sup>20</sup> The direct consequence of such coupling would be observation of a superfluid response due to weak Meissner effect associated with Josephson current. Thus, the model of Josephson coupled layers is able to reconcile the nonmetallic normal state properties along the  $c$ -axis with the evidence for superconducting penetration depth at  $T < T_c$  (Fig. 4).

As shown in Ref. 27 the in-plane penetration depth of an array of weakly coupled superconducting layers is given by

$$c^2/\lambda_{\parallel}^2 = (4\pi n_s e^2)/m^*, \quad (2)$$

where  $n_s$  is concentration of superconducting carriers and  $m^*$  is effective mass. The out-of-plane value of the penetration depth is

$$\lambda_{\perp}^2 = \hbar c^2/8\pi d e J_c, \quad (3)$$

where  $d$  is distance between planes and  $J_c$  is the density of Josephson current between the planes. Within BCS theory the value of  $J_c$  can be related to the normal state resistance  $R_n$  and the energy gap as<sup>28</sup>

$$J_c = \pi\Delta(T) \tanh[\Delta(T)/2kT]/2eR_n. \quad (4)$$

The important difference between  $\lambda_{\parallel}$  and  $\lambda_{\perp}$  is that the former is determined solely by the superfluid density  $n_s/m^*$  whereas the latter is governed by the probability of the interplane tunneling expressed in terms of  $J_c$ . Thus, one expects to observe relatively small variation of  $\lambda_{\parallel}$  in different high- $T_c$  materials. Indeed, the in-plane penetration depth in all crystals shown in Fig. 4 is between 800 and 2750 Å.<sup>29</sup> As for the out-of-plane penetration depth, it scales with the  $c$ -axis conductivity  $\sigma_1 = d/R_n$  and thus the variation of  $\lambda_{\perp}$  may be of several orders of magnitude, in agreement with the experimental results plotted in Fig. 4. The absolute value of  $\lambda_{\perp}$  is strongly enhanced compared to the in-plane value since the density of Josephson current is quite small, also consistent with data.

Combining Eq. (3) and Eq. (4) we obtain Eq. (1) for the out-of-plane penetration depth  $\lambda_{\perp}$ . Thus, the formal description of the penetration depth in terms of the normal state conductivity and the gap appears to be the same for the penetration depth of a dirty limit superconductor and of  $\lambda_{\perp}$  of a layered superconductor with Josephson coupling. Therefore both models describe with equal success the correlation between  $\lambda_c$  and the  $c$ -axis conductivity plotted in Fig. 4. In our view the advantage of the Josephson coupling approach is that it (a) naturally explains the anisotropy of the penetration depth in crystals of high- $T_c$  materials, (b) is not restricted to

the  $c$ -axis conductivity being in the metallic regime, (c) is related to the well-defined layered structure of cuprates and the anisotropy of their normal state properties. Finally, evidence for Josephson coupling is provided by *direct* experiments on the single crystals of  $\text{Bi}_2\text{Ca}_2\text{SrCu}_2\text{O}_8$ .<sup>20</sup> This suggests that the superconducting properties along the  $c$  axis can be understood in terms of Josephson coupling between the  $\text{CuO}_2$  planes.

A better approximation than given by Eqs. (1)–(4) would be to allow for possible gap anisotropy. We believe that relatively small values of  $2\Delta = 128 \text{ cm}^{-1}$  and  $320 \text{ cm}^{-1}$  required to reproduce the correlation in Fig. 4 may indicate that it is the *average* gap in the  $ab$ -plane which is important for the Josephson tunneling along the  $c$  axis. It is not clear at the moment how the particular symmetry of the gap function may affect the interplane tunneling. The case of  $d$ -wave gap needs special consideration because the matrix element for the incoherent pair tunneling may be equal to zero.<sup>30</sup> In the absence of detailed knowledge of gap anisotropy, the extraction of a reliable value of the gap from the  $c$ -axis penetration depth data would be premature.

The correlation between the strength of the interplane coupling described by the quantities,  $\sigma_1$  and  $\lambda_c$ , revealed in Fig. 4, indicates that there is no correlation between interplane coupling and the value of the critical temperature in high- $T_c$  materials. This is clearly illustrated by the fact that  $\text{Bi}_2\text{Ca}_2\text{SrCu}_2\text{O}_8$  which has the lowest value of the  $c$  axis conductivity and the highest value of  $\lambda_c$  shows nearly the same critical temperature as  $\text{YBa}_2\text{Cu}_4\text{O}_8$  which is the oppo-

site extreme case with nearly highest conductivity and the lowest penetration depth.

In conclusion, we have observed evidence for a pseudogap in the  $c$ -axis conductivity of  $\text{YBa}_2\text{Cu}_4\text{O}_8$  single crystal similar to that seen in the oxygen deficient  $\text{YBa}_2\text{Cu}_3\text{O}_{6.70}$  material. While the pseudogap cannot be attributed to a conventional gap in the spectrum of charge excitations, its assignment to a spin gap seems to be quite plausible as indicated by FIR spectroscopy, NMR and transport data. We found a correlation between the value of the  $c$ -axis penetration depth and the conductivity perpendicular to the  $\text{CuO}_2$  layers in a variety of high- $T_c$  superconductors. The possibilities of dirty limit superconductivity and of Josephson coupling between  $\text{CuO}_2$  layers are discussed in the context of the superconducting state anisotropy of various cuprates.

*Note added.* In a recent paper by Shibauchi *et al.*<sup>31</sup> the authors carry out an analysis similar to the one presented here for the crystals of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .

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<sup>29</sup>Our analysis of the  $a$ - and  $b$ -axes conductivities in the crystals  $\text{YBa}_2\text{Cu}_4\text{O}_8$  from the same batch as those used for the  $c$ -axis study yields the following values of the penetration depth:  $\lambda_a = 2000 \text{ \AA}$ ,  $\lambda_b = 800 \text{ \AA}$ . For the optimally doped  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ ,  $\lambda_{ab} = 1440 \text{ \AA}$  (Ref. 32); for  $\text{Pb}_2\text{Sr}_2\text{RCu}_3\text{O}_8$ ,  $\lambda_{ab} = 2575 \text{ \AA}$  (Ref. 23); for ( $\text{La}_{1.84}\text{Sr}_{0.16}\text{CuO}_4$ ),  $\lambda_{ab} = 2750 \text{ \AA}$  (Ref. 33); for  $\text{Bi}_2\text{Ca}_2\text{SrCu}_2\text{O}_8$ ,  $\lambda_{ab} = 1800 \text{ \AA}$  (Ref. 34).

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