PHYSICAL REVIEW B VOLUME 38, NUMBER 1 1 JULY 1988

# Possibility of an increase in density of states to explain the $T_c$ change from $La_{2-x}Sr_xCuO_4$ to $YBa_2Cu_3O_7$

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(Received 5 January 1988; revised manuscript received 31 March 1988)

Within a simple and natural tight-binding model, we show that the increase in critical temperature from $La_{2-x}Sr_xCuO_4$ (or $YBa_2Cu_3O_{7-\delta}$ with $\delta \approx 0.5$) to $YBa_2Cu_3O_7$ can easily be explained by the increase in density of states due to the CuO chains present in $YBa_2Cu_3O_7$. We discuss the effects of moving the Fermi energy and shifting the chain band with respect to the plane bands.

## I. INTRODUCTION

There is no agreement to date on why the $T_c$ of the superconductors $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_7$ is so high.[1,2] The major problem is that the most distinctive property of these compounds is the high value of their $T_c$ and directly related properties. Unfortunately theory has found it very hard to predict critical temperatures, therefore it is not easy to reach definite conclusions. An impressive number of explanations have been presented,[2] most of them proposing nonphonon mechanisms. When the standard phonon-mediated superconductivity is investigated, the usual conclusion is that a rather strong electron-phonon coupling is necessary to explain the high $T_c$.

In this paper we add to the confusion by pursuing a third line of thought, namely that the high $T_c$ might be due to the occurrence of a fairly large density of states around the Fermi level. This idea has already been proposed for transition metals and $A15$ compounds[3] where the quasi-one-dimensional structure due to linear chains of atoms leads to a high density of states at the edge of the corresponding bands. In a similar way it has been suggested[4] that the high density of states due to the Van Hove singularities in quasibidimensional systems might lead to an increase in $T_c$. This proposal has been taken up[5] to explain the high $T_c$ of $La_{2-x}Sr_xCuO_4$ since these compounds display a fairly strong bidimensional behavior. It is important to note that a large density of states does not exclude a nonphonon mechanism nor a strong-coupling superconductor.

Now the discovery of $T_c \approx 95$ K in $YBa_2Cu_3O_7$ raises the question, is it possible to account for this huge change in $T_c$ by an increase in density of states alone? Naturally this is always possible if we take an *ad hoc* increase in density of states. The relevant question is to known if the required increase is compatible with our present knowledge on $YBa_2Cu_3O_7$. In this paper we investigate this problem within the simplest possible model: tight-binding band-structure, weak-coupling Bardeen-Cooper-Schrieffer (BCS) theory. An essential feature of our paper is that we do not calculate $T_c$, but we rather try to *compare* the $T_c$'s in $La_{2-x}Sr_xCuO_4$ and in $YBa_2Cu_3O_7$ by assuming that only the density of states changes while the cutoff frequency and the strength of the electron-electron interaction are unchanged. In this way we avoid the notoriously uncertain evaluation of this interaction in the calculation of $T_c$. Or, in other words, we extract the interaction from the $T_c$ of one compound to calculate the $T_c$ of the other one. Our conclusion is that we can account easily for the change in critical temperature.

Naturally it is known that there are strong correlations in these compounds. Correlations are already strong in any ordinary metal. Here they are expected to be stronger due to the rather narrow bands involved in $YBa_2Cu_3O_7$. The correlations are clearly shown experimentally by antiferromagnetism of the related compounds $La_2CuO_4$ and $YBa_2Cu_3O_6$. Our approach to this problem is what is usually referred to as the weak-coupling approach. We assume that correlations lead to a renormalization of the low-lying excitations and that the correct physical picture is a Fermi liquid with standard metallic behavior. To be proper we should rephrase our paper and speak in terms of quasiparticles rather than electrons. Within this approach the antiferromagnetism is explained by a spin-density wave favored by a strong nesting of the Fermi surface, superconductivity appearing when the Fermi level is away from these particular values. Naturally the weak-coupling approach may be inadequate for the high-$T_c$ compounds. It is likely that this question will first be answered by experiment.

Having said all this, we are faced with the fact that we do not know the renormalized quantities we need to obtain $T_c$. Our approach is to take the simplest model for the excitation spectrum of our compounds and extract the single parameter we need from band-structure calculations, which give bare quantities. This would not be reliable if we wanted to calculate $T_c$. But we only want to show that the *possibility* that the increase in $T_c$ results from an increase in density of states *cannot be dismissed.* Therefore taking the bare quantities is a reasonable procedure because if the possibility exists with bare quantities, it cannot be rejected with dressed quantities. Let us finally note that interactions are likely to lead to heavier effective masses and higher density of states and therefore to reinforce the effects we consider.

## II. THE MODEL

$YBa_2Cu_3O_7$ can be considered[6] as being made up of two layers of $CuO_2$ square lattices separated by a layer of parallel CuO chains (we neglect the small orthorhombic distorsion). The CuO distance is very near $a/2=1.93$ Å. The $CuO_2$ planes are separated from the CuO chains by BaO planes. Finally this whole structure is separated from the next one by a layer of Y atoms.

The $CuO_2$ planes are essentially identical to the ones found in $La_2CuO_4$ (the CuO distance is rather 1.90 Å but we neglect this small change). In this last compound these planes are separated by two LaO layers. In $La_2CuO_4$ the conduction band can be described by a tight-binding approximation.[5,7] The electron hops from one Cu atom to the next using the oxygen orbitals as intermediate state. This corresponds to a hybridization of Cu $d_{x^2-y^2}$ orbitals with O $p_x$ and O $p_y$ orbitals. Since chemically we must have a $Cu^{2+}$ valence, the Fermi level is in the antibonding band. With only nearest-neighbor terms, the dispersion relation reads

$$\varepsilon(\mathbf{k})=2t_1[2-\cos(k_xa)-\cos(k_ya)]\,, \tag{1}$$

if we assume that the transfer integral $\gamma_1$ between O and Cu is small compared to the difference $E_d-E_p$ between the atomic orbital energies. The energy $E_d$ of the Cu orbital is taken as zero. A perturbation calculation gives $t_1=\gamma_1^2/(E_d-E_p)$ for the effective transfer integral $t_1$ between two Cu atoms. This simple tight-binding approximation is actually in rather good agreement with detailed band-structure calculations[8,9] which typically give $t_1=0.5$ eV leading to full bandwidth of 4 eV. Since the $CuO_2$ planes in $YBa_2Cu_3O_7$ are essentially identical to those of $La_2CuO_4$, we take the same band structure given by Eq. (1).

A similar tight-binding approximation for the one-dimensional CuO chains gives

$$\varepsilon(\mathbf{k})=2t_2[1-\cos(k_xa)]\,, \tag{2}$$

where the effective transfer integral $t_2$ between two Cu atoms should not be very different from $t_1$ since the CuO distances are the same in the planes and in the chains. However, in this way we forget the effect of the O atoms of the BaO planes which are located just above and below the Cu atoms of the chains at a distance of 1.85 Å. These are easily taken into account in a tight-binding calculation, but in the limit of small transfer integral $\gamma_3$ these two atoms together merely shift the energy of the Cu orbital by $2\gamma_3^2/(E_d-E_p)=2t_3$ where $t_3$ should be somewhat larger than $t_1$ since $\gamma_3$ is increased by perhaps 10% with respect to $\gamma_1$ due to the 0.08-Å reduction in the CuO distance. Therefore Eq. (2) is replaced by

$$\varepsilon(\mathbf{k})=2t_2[1-\cos(k_xa)]+2t_3\,. \tag{3}$$

Now the plane band Eq. (1) has a Van Hove singularity at the middle of the band $\varepsilon=4t_1$ with a logarithmic divergence while the chain band Eq. (3) has a singularity at its bottom with an inverse square-root divergence. Both singularities provide an increase in *average* density of states which is favorable for a high $T_c$. We note that these two singularities are not far from each other, the energy difference being $\varepsilon_0=4t_1-2t_3$. This difference is likely to be even reduced since the Coulomb repulsion due to the O atoms of the BaO plane is stronger on the Cu of the chains than the corresponding repulsion on the Cu of the planes because of the smaller distance (1.85 Å against 1.93 Å).

Anyway, *ab initio* band-structure calculations have already been performed[10-14] for $YBa_2Cu_3O_7$ and they all find that the bottom of the chain band is very near (from 0.1 to 0.3 eV) below the middle of the plane bands (which we identify as being 2 eV below the top; the lower part of the band seems rather complicated due to various couplings but this is unimportant since this happens far from the Fermi level). Therefore, we are very near an ideal situation where the bottom of the chain band is just located at the middle of the two plane bands.[15]

Now by balancing the valence numbers, the three Cu atoms of a cell must carry seven holes, four of them being in the *d* bands. In the ideal situation this is obtained by putting two holes in the chain band and one hole in each of the plane bands. Therefore, the chain band is empty, the plane bands are half filled, and the Fermi level falls right at the bottom of the chain band and in the middle of the plane bands. Thus, it is located at the same place as the two above singularities, a situation which could explain the high $T_c$ of $YBa_2Cu_3O_7$. Band-structure calculations find the Fermi level very near this ideal position (the rapidly varying density of states might explain the notable differences obtained for its value at the Fermi level by the various calculations).

Naturally, such a remarkable ideal situation cannot be exact and indeed band-structure calculations, within their precision, find only a near coincidence. Moreover, there can actually be no divergences in the density of states. They occur only because we have assumed pure one- and two-dimensional behavior which is actually spoiled by coupling between the various parts of the system. However, we do have interchain coupling but this should be small because of the large distance (3.86 Å) between the Cu belonging to neighboring chains. In the same way the chains are coupled to the planes through the O atoms of the BaO planes. But the $p_z$ orbital of this O atom, which is strongly coupled to the chains as we have seen, has for symmetry reasons a zero overlap integral with the $d_{x^2-y^2}$ orbital of the Cu atoms belonging to the planes, which is the important one for the planes. This O $p_z$ orbital has a strong overlap with the Cu$d_{3z^2-r^2}$ orbital of the same atoms but these orbitals have a small weight at the Fermi level in the planes. Therefore, the coupling between the chains and the planes are weak near the Fermi level. Finally, the $CuO_2$ planes on both sides of the Y layer should also be badly coupled because of the large distance (3.33 Å) between corresponding Cu atoms and the fact that Y atoms offer a poor relay. A similar situation arises in $La_{2-x}Sr_xCuO_4$. Naturally the question is to know exactly how small these couplings are, but a result of order 0.1 eV would be enough for our purpose, which is not unreasonable since it would be 20% of our parameter *t*. Now the band-structure calculations all find quasi-two-dimensional $CuO_2$ planes and quasi-one-dimensional CuO chains behavior near the Fermi level. Therefore, our model can be considered as retaining the salient features[16] of the *ab initio* band-structure calculations.

Regarding this smearing problem, it is very important to realize that a broadening of the density of states over typically a full width of 4 $k_B T_c$ has essentially no effect on $T_c$, and densities of states beyond that range still have an important weight in the value of $T_c$. This means that a number of broadening effects, which are dramatic for a $T_c \approx 10$ K superconductor, become inessential for a $T_c \approx 95$ K superconductor. In other words, high-$T_c$ superconductivity helps high-$T_c$ superconductivity. This is a curious bootstrap effect, and this is perhaps of some relevance for the remarkable $T_c$ stability and insensitivity of the 95-K superconductor.

In this paper, as a means to compare $YBa_2Cu_3O_7$ and $La_{2-x}Sr_xCuO_4$, we want to study the change in $T_c$ for $YBa_2Cu_3O_{7-\delta}$ as a function of the departure $\delta$ from oxygen stoichiometry. There are already experimental results[17,18] for this dependence and we will consider them in the discussion of our results. Neutron-diffraction experiments[18] show that the missing oxygen is removed from the CuO chains (there is also the possibility of disordering when an O atom is between two Cu belonging to neighboring chains but we will ignore it.) Now when an O atom is removed from a chain, the Coulomb repulsion due to this atom and felt by the two $d_{x^2-z^2}$ orbitals of the neighbor Cu atoms disappears. Therefore, the energy of these two orbitals are reduced by probably several eV, giving rise to states with energy much below the chain band (these are similar to dangling bonds). The natural and most simple hypothesis is to assume that these states are localized. These two states are now filled whereas they were empty in the presence of the O atom. But when this O atom disappears, we have to find room for two electrons which were previously accommodated by the O atom, giving an $O^{2-}$ ion. These two electrons will merely go in the two localized states and we obtain two $Cu^{2+}$ instead of the two $Cu^{3+}$ we had before the O removal (naturally these simple valences do not correspond to reality, they are just convenient for easy counting). In summary, we go from $Cu^{3+}O^{2-}Cu^{3+}$ to $Cu^{2+}$-$Cu^{2+}$ when one O atom is removed.

Now these localized electrons (we should speak rather of localized holes) should produce paramagnetic moments. Actually such a paramagnetic susceptibility, increasing with the nonstoichiometry $\delta$, has already been observed[18] in $YBa_2Cu_3O_{7-\delta}$, but the magnitude of this susceptibility seems to be too low for our purpose. One possibility is that the two neighboring $Cu^{2+}$ interact antiferromagnetically, giving rise to a nonmagnetic singlet state. Another one is O vacancy interactions which could conceivably reduce the paramagnetic behavior. It is also possible that, due to hybridization with other low-energy $d$ orbitals, our states are somewhat delocalized leading to reduction of the magnetic moment. But it might well be that these states are so delocalized that we can put two electrons of opposite spins in these states, leading effectively to $Cu^+$. Actually, the whole question of the physical nature of these states is an open problem which looks quite difficult to solve theoretically. Therefore, we will stick to the simple hypothesis of localized states for simplicity. Let us just mention that other hypotheses would not qualitatively modify our basic conclusion. But it would complicate the calculations and obviously change the results quantitatively.

The net result of our hypothesis is that the Fermi level is unchanged when an O atom is removed from the chains since the two electrons left by this atom find a place in new deep lying states and not at the Fermi level. Naturally, the chain band is strongly affected by the oxygen removal. A similar situation[19] has been studied for the $T_c$ dependence of $V_3$ Au on atomic ordering. Here we make use of the following simple version of this study. For a nonstoichiometry $\delta$, one O atom among $1/\delta$ is removed which leaves, on the average, chains made of $1/\delta$ Cu atoms. The lowest possible wave vector $k_0$ for a wave function corresponds to half a wavelength in the chain which gives $k_0 = \pi\delta/a$. We cut off the spectrum at this wave vector assuming that the lower energy part of the spectrum, corresponding to $k < k_0$, has disappeared because of the oxygen removal. This leads to a rapid disappearance of the essential part of the chain band spectrum for the critical temperature and a corresponding drop in $T_c$ which is observed experimentally. Rather soon the chain band plays no role and we are basically left only with $CuO_2$ planes. This is essentially the situation found in $La_{2-x}Sr_xCuO_4$. Indeed, experiments[17,18] show that $T_c$ falls in the vicinity of 40 K for $YBa_2Cu_3O_{7-\delta}$ with $\delta \approx 0.5$ and for larger $\delta$ superconductivity disappears completely. This temperature is rather near the typical $T_c$ for $La_{2-x}Sr_xCuO_4$. Therefore, it is reasonable to ascribe the difference between the $T_c = 95$ K of $YBa_2Cu_3O_7$ and the $T_c \approx 40$ K of $YBa_2Cu_3O_{7-\delta}$ with $\delta \approx 0.5$ (or equivalently $La_{2-x}Sr_xCuO_4$) to the disappearance of the chain band.[20]

Actually, very recent experiments show that $YBa_2Cu_3O_6$ is antiferromagnetic. This can be understood by assuming that further oxygen removal leads to $Cu^+$ in the "chains." Then we are left in $YBa_2Cu_3O_6$ with half-filled $CuO_2$ plane bands, a situation identical to $La_2CuO_4$ where antiferromagnetism is also found. This makes it likely that $YBa_2Cu_3O_7$ does not correspond to the ideal situation since this might also lead to antiferromagnetism. Rather a Fermi level located 0.1 eV above the bottom of the chain band (more in agreement with band-structure calculations) would put the plane band (we would have 2.3 holes in the plane band which corresponds to the situation found in $La_{2-x}Sr_xCuO_4$ with $x = 0.15$) away enough from half filling to avoid this instability. The position with respect to the Fermi level of the Van Hove singularity of the plane bands plays a rather secondary role in our model for the calculation of the variation of $T_c$ (see below). In the real $YBa_2Cu_3O_7$ there is no reason to have it located at the middle of the plane band. So in the following, we will discuss our results as a function of its location. We stress that the essential ingredient of our model is the bottom of the chain band located not far away from the Fermi level.

In the following, we quantitatively study the model corresponding to the band structure, Eqs. (1) and (3), and look for the change in $T_c$ when the chain band Eq. (3) disappears. We consider, in particular, the effect on this change of shifting the Fermi energy and of moving the chain band with respect to the plane bands. Although we have explained that the Van Hove singularities are actually smeared out, we will keep for mathematical convenience the singular density of states corresponding to Eqs. (1) and (3). The reason is that the BCS gap equation itself produces an averaging over typically $4k_BT_c$, and therefore any effect of the singularities is completely washed out. Thus, keeping the singular behavior is actually unimportant, and since it is convenient for calculation, we keep it. We could cut off the singularities. This would complicate the results unnecessarily and bring little quantitative changes.

## III. CALCULATION OF THE $T_c$ CHANGE AND DISCUSSION

The density of states per spin corresponding to the plane band Eq. (1) is given for $\epsilon < 4t_1$ by

$$N_1(\epsilon)=\frac{4N}{\pi^2}\frac{1}{8t_1-\epsilon}K\left[\frac{\epsilon}{8t_1-\epsilon}\right] , \tag{4}$$

where $K$ is an elliptic integral[21] and $N$ the number of Cu atoms in the plane. In the vicinity of the middle of the band $\epsilon\approx 4t_1$, $N_1(\epsilon)$ can be approximated by[19]

$$N_1(\epsilon)\approx\frac{N}{2\pi^2 t_1}\ln\left[\frac{16t_1}{|\epsilon-4t_1|}\right] , \tag{5}$$

the corrective terms[22] being of order $(\epsilon-4t_1)\ln|\epsilon-4t_1|$.

The density of states for the chain band Eq. (3) is, with $\epsilon'=\epsilon-2(t_2+t_3)$

$$N_2(\epsilon)=\frac{N}{2\pi t_2}\left[1-\left[\frac{\epsilon'}{2t_2}\right]^2\right]^{-1/2}\approx\frac{N}{2\pi t_2}\left[\frac{\epsilon'+2t_2}{t_2}\right]^{-1/2} , \tag{6}$$

where the last equality holds near the bottom of the band $\epsilon'\approx -2t_2$.

The BCS gap equation reads

$$\frac{2}{V}=\int_{-\omega_0}^{\omega_0}d\xi\frac{\tanh(\xi/2k_BT_c)}{\xi}N(\xi) , \tag{7}$$

where $\xi=\epsilon-E_F$ is the energy measured from the Fermi level and $\omega_0$ is the BCS cutoff. The density of states $N(\xi)$ is twice $N_1$ corresponding to the two plane bands plus $N_2$ corresponding to the chain band. We measure the energy from the middle of the plane band. It is also convenient to work with reduced energies taking $2k_BT_C$ as unity. We set

$$\begin{gathered}\epsilon-4t_1=2k_BT_cy,\ \ E_F=\epsilon_F-4t_1=2k_BT_cy_F ,\\ \omega_0=2k_BT_c\Omega,\ \ t_1=2k_BT_cb ,\\ \epsilon_0=2t_3-4t_1=2k_BT_cy_0 ,\end{gathered} \tag{8}$$

and the gap equation becomes

$$\frac{2\pi^2t_1}{NV}=I=\int_{-\Omega+y_F}^{\Omega+y_F}dy\frac{\tanh(y-y_F)}{y-y_F}\left[\ln\left(\frac{b}{|y|}\right)+v_1+v_2\left(\frac{b}{y-y_0}\right)^{1/2}\right] , \tag{9}$$

where in our simple model

$$v_1=4\ln 2,\ \ v_2=\frac{\pi}{2}\left(\frac{t_1}{t_2}\right)^{1/2} . \tag{10}$$

In the following we take $t_2=t_1$. We treat first the ideal situation $y_F=y_0=0$ where the two singularities coincide with the Fermi level and then consider the effect of the departure from this ideal situation. The calculation corresponding to the first two terms within brackets in Eq. (9) is carried out through a by part integration

$$\begin{aligned}\int_{-\Omega}^{\Omega}dy\frac{\tanh y}{y}\ln\frac{be^{v_1}}{|y|}&=2\ln(be^{v_1})\ln\Omega\\ &\quad-\ln^2\Omega+2\alpha\ln(be^{v_1})+\beta ,\end{aligned} \tag{11}$$

where

$$\alpha=-\int_0^{\infty}dy\frac{\ln y}{\cosh^2 y},\ \ \beta=\int_0^{\infty}dy\frac{\ln^2 y}{\cosh^2 y} . \tag{12}$$

In obtaining Eqs. (11) and (12) we have replaced the boundaries $\pm\,\Omega$ in the integrals for $\alpha$ and $\beta$ by $\pm\,\infty$ and we have made $\tanh\Omega\approx 1$. Both of these approximations produce only exponentially small errors of order $e^{-2\Omega}$. The integral $\alpha$ is well known in BCS theory $\alpha=\gamma-\ln(\pi/4)\approx 0.818$ where $\gamma$ is the Euler constant ($e^{\alpha}/2=1.13$) and numerically $\beta\approx 1.99$. Assuming stoichiometry ($\delta=0$) the last term in Eq. (9) gives for $YBa_2Cu_3O_7$

$$\int_0^{\Omega}dy\frac{\tanh y}{y_{3/2}}=L(0)-\int_{\Omega}^{\infty}dy\frac{\tanh y}{y^{3/2}}=L(0)-2(\Omega)^{-1/2} , \tag{13}$$

with again exponentially small errors and

$$L(0)=\int_0^{\infty}dy\frac{\tanh y}{y^{3/2}}=\frac{4}{\sqrt{\pi}}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^{3/2}}\approx 3.81 , \tag{14}$$

where the series is obtained by residues calculation (see below). Finally, Eq. (9) reads for $YBa_2Cu_3O_7$

$$\begin{aligned}I=&\ln^2(b_0e^{\alpha+v_1})-\ln^2(r_0e^{v_1})\\ &+v_2b_0^{1/2}L(0)-2v_2r_0^{1/2}+\beta-\alpha^2 ,\end{aligned} \tag{15}$$

with $r_0=t_1/\omega_0=b/\Omega$ and we have set $b_0=t_1/2k_BT_c^0$ where $T_c^0$ is the critical temperature of $YBa_2Cu_3O_7$.

Now for $YBa_2Cu_3O_{7-\delta}$, the lower boundary in the integral Eq. (13) must be replaced by $c=\pi^2\delta^2bt_2/t_1$ corresponding to a cutoff energy $\Delta\epsilon_0=t_2(k_0a)^2$ due to oxygen depletion. Taking the difference between Eq. (15) and the corresponding equation for $c\neq 0$ we obtain

$$\frac{1}{\nu_2\sqrt{b}}[\ln^2(be^{\alpha+\nu_1})-\ln^2(b_0e^{\alpha+\nu_1})] \\ +L(0)\left[1-\left(\frac{b_0}{b}\right)^{1/2}\right]=\int_0^c dy\frac{\tanh y}{y^{3/2}}\ , \quad (16)$$

which gives the critical temperature $k_BT_c=t_1/2b$ of $YBa_2Cu_3O_{7-\delta}$.

Let us first consider the limit case where the chain band has disappeared. This corresponds to taking $c=\Omega$ and Eq. (16) reduces to

$$\ln^2(b_1e^{\alpha+\nu_1}-\ln^2(b_0e^{\alpha+\nu_1})=L(0)\nu_2b_0^{1/2}-2\nu_2r_0^{1/2}\ , \quad (17)$$

where $b_1=t_1/2k_BT_c^1$ and $T_c^1$ is the critical temperature of $YBa_2Cu_3O_{7-\delta}$ for $\delta\approx 0.5$ or equivalently of $La_{2-x}Sr_xCuO_4$ as we discussed. If we take for $\nu_1$ and $\nu_2$ the values given by our simple model Eq. (10) with $t_2=t_1$, everything is known in Eq. (17) except $r_0$. We take for example $\omega_0=0.05$ eV, a typical high-energy phonon in these compounds. With $t_1=0.5$ eV this gives $r_0=10$. Then from Eq. (17) $T_c^0=95$ K leads to $T_c^1=21$ K, much too low compared to the expected 40 K. Conversely $T_c^1=40$ K leads to the much too high $T_c^0=135$ K. These results are not very sensitive to our assumed value for $r_0$. Therefore we come to the conclusion that the change in density of states alone can easily explain the observed value for the ratio $T_c^0/T_c^1$. Since we have considered an ideal situation, it is not surprising that we come with a much too high $T_c^0/T_c^1$. All the differences between the real situation and the ideal one will reduce this ratio, leading to a possible agreement with experiment.

We now consider the $\delta$ dependence of the critical temperature. We see immediately that our limiting temperature $T_c^1$ is reached for $c=\Omega$, that is $\delta=(\omega_0/t_2)^{1/2}/\pi\approx 0.1$. For smaller $\delta$, the decrease in $T_c$ is linear in $\delta$ and slows down somewhat near $\delta=0.1$. This rapid drop in $T_c$ is in sharp contrast with the experimental results[17,18] where $T_c$ has not changed very much for $\delta=0.1$ and the rapid change from 90 to 50 K takes place between $\delta=0.1$ and $\delta=0.4$. Clearly our rapid drop is partly due to the location at the Fermi level of the low-energy part of the spectrum, which is sensitive to oxygen depletion. The situation would be much improved, and much more similar to experiment, if we were to place the bottom of the chain band at say 0.1 eV below the Fermi level ($\varepsilon_0=-0.1$ eV), more in agreement with band-structure calculations. Indeed for $\delta<0.1$ the affected part of the spectrum would be outside $[-\omega_0,\omega_0]$: $T_c$ would only begin to drop at $\delta=0.1$ and it would decrease more gently than in the ideal situation. Also if instead of a sharp cutoff $k_0$ we used a more careful treatment[19] of the effect of oxygen removal on the spectrum, we would obtain a less drastic effect. We would also much improve the situation by choosing for the cutoff $\omega_0$ a more reasonable value $\omega_0=0.1$ eV corresponding to the highest phonon frequency. All this would bring us close to the experimental results.

However, we have not proceeded to a quantitative study because at the present stage the physical interpretation of the experimental data is not so clear. Indeed the more recent data[18] seems to show a kind of plateau between say $\delta=0.25$ and $\delta=0.4$ with $T_c\approx 55$ K which requires interpretation. It is likely that what happens is more complicated than a simple homogeneous oxygen removal. The plateau suggests the presence of yet another unidentified superconducting phase which is perhaps an ordered compound. In a similar way one might think that the stability[18] of $T_c$ at 90 K is due to inhomogeneous effects, oxygen being removed from some regions while others stay at the stoichiometric $YBa_2Cu_3O_7$. Pending the solution of these experimental questions, we cannot obtain definite conclusions from a comparison between experiment and our model. For example, one could imagine[23] that the $T_c$ of $YBa_2Cu_3O_7$ is reduced by some small disorder and the ordered $YBa_2Cu_3O_7$ would have a much higher $T_c$, which would experimentally give a much more rapid drop of $T_c$ with $\delta$.

Let us turn to the effect of shifting the Fermi level and the bottom of the chain band from their ideal position at the middle of the plane band.[24] The contribution of the constant part of the density of states $\nu_1+\ln b$ in Eq. (9) is unchanged when $y_F\neq 0$. The contribution of the $\ln|y|$ term is changed by

$$\int_{-\Omega+y_F}^{\Omega+y_F}dy\frac{\tanh(y-y_F)}{y-y_F}\ln\frac{1}{|y|}-\int_{-\Omega}^{\Omega}dy\frac{\tanh y}{y}\ln\frac{1}{|y|}=-J(y_F)+\int_{\Omega}^{\infty}\frac{dy}{y}\ln\left|1-\frac{y_F^2}{y^2}\right|\ , \quad (18)$$

where

$$J(y_F)=\int_{-\infty}^{+\infty}dy\frac{\tanh y}{y}\ln|1+y_F/y|\ . \quad (19)$$

The second term on the right-hand side of Eq. (18) can be expanded in powers of $y_F/\Omega$

$$\int_{\Omega}^{\infty}\frac{dy}{y}\ln\left(1-\frac{y_F^2}{y^2}\right)=-\frac{1}{2}\sum_1^{\infty}\frac{1}{n^2}\left(\frac{y_F}{\Omega}\right)^{2n}\ , \quad (20)$$

which takes for $y_F=\Omega$ the value $-\pi^2/12$. $J(y_F)$ can be integrated by residues as

$$J(y_F)=2\sum_{n=0}^{\infty}\frac{1}{2n+1}\ln\left(1+\frac{4}{\pi^2}\frac{y_F^2}{(2n+1)^2}\right)\ . \quad (21)$$

When an expansion in powers of $y_F^2$ is performed one sees that the $n=0$ by far dominates except for the lower powers of $y_F^2$. If correction is made for this point one obtains a very good approximation for $J(y_F)$. For example,

$$J(y_F)=2\ln\left(1+\frac{4y_F^2}{\pi^2}\right)+4.2y_F^2 10^{-2}-7.43y_F^4 10^{-4}\ , \quad (22)$$

gives a very good approximation with errors of order $10^{-2}$ up to $y_F = 3$ where $J(y_F) = 3.39$. As can be seen directly from Eq. (19) for large $y_F$ the correct behavior is actually

$$J(y_F) \approx \ln^2 y_F + 2a \ln y_F + \beta - \frac{\pi^2}{6} + \frac{\pi^2}{24 y_F^2} , \tag{23}$$

which is already a very good approximation for $y_F = 3$ and even rather good for $y_F = 1$.

Similarly the last term in Eq. (9) gives (for $c = 0$) a contribution

$$\begin{aligned} F(Y_F) &= \int_0^{\Omega + Y_F} \frac{dY}{\sqrt{Y}} \frac{\tanh(Y - Y_F)}{Y - Y_F} \\ &= L(Y_F) - \int_{\Omega + Y_F}^{\infty} \frac{dY}{\sqrt{Y}} \frac{1}{Y - Y_F} \end{aligned} \tag{24}$$

where we have set $Y_F = y_F - y_0$ and assume $|Y_F| < \Omega$. The last term on the right-hand side of Eq. (24) is

$$\begin{aligned} &\frac{2}{\sqrt{Y_F}} \ln \left[ \left( 1 + \frac{Y_F}{\Omega} \right)^{1/2} + \left( \frac{Y_F}{\Omega} \right)^{1/2} \right], \quad Y_F > 0 , \\ &\frac{1}{\sqrt{|Y_F|}} \left[ \pi - 2 \arctan \left( \frac{\Omega + Y_F}{|Y_F|} \right)^{1/2} \right], \quad Y_F < 0 . \end{aligned} \tag{25}$$

In the case $Y_F > \Omega$, one must also subtract from the right-hand side of Eq. (24)

$$\int_0^{Y_F - \Omega} \frac{dY}{\sqrt{Y}} \frac{\tanh(Y - Y_F)}{Y - Y_F} = \frac{2}{\sqrt{Y_F}} \ln \left[ \left( \frac{Y_F}{\Omega} - 1 \right)^{1/2} + \left( \frac{Y_F}{\Omega} \right)^{1/2} \right] . \tag{26}$$

The first term on the right-hand side of Eq. (24) can be integrated by residues as

$$\begin{aligned} L(Y_F) &= \int_0^{\infty} \frac{dY}{\sqrt{Y}} \frac{\tanh(Y - Y_F)}{Y - Y_F} \\ &= 2\sqrt{2} \sum_{n=0}^{\infty} \frac{1}{2n+1} \frac{\sqrt{Y_F + g(Y_F, n)}}{g(Y_F, n)} , \end{aligned} \tag{27}$$

where $g(Y_F, n) = [Y_F^2 + \pi^2 (2n+1)^2/4]^{1/2}$. Again a good approximation is obtained with the term $n = 0$, corrected to give the exact result for the lower powers of the expansion in powers of $Y_F$. For large $Y_F > 0$

$$L(Y_F) \approx \frac{2}{\sqrt{Y_F}} [a + 2 \ln(2\sqrt{Y_F})] , \tag{28}$$

which is already good for $Y_F = 3$. And for large $Y_F < 0$

$$L(Y_F) \approx \frac{\pi}{\sqrt{|Y_F|}} , \tag{29}$$

which is a good approximation at $Y_F = -2$. We have $L(0) = 3.81$ and $L(Y_F)$ has a maximum value 4.19 very near $Y_F = 1$. Then it decreases rather slowly since $L(5) = 3.65$. We see that the ideal situation $Y_F = 0$ is not the optimal one for $L(Y_F)$ which is actually obtained for $Y_F \approx 1$. We note that $L(Y_F)$ does not change so much for positive values of $Y_F$ (provided $Y_F < \Omega$). For example, if the bottom of the chain band is 0.05 eV below the Fermi level we will have $Y_F = 2.9$ and $L(2.9) \approx 3.81 \approx L(0)$. On the other hand the situation is obviously much less favorable when the bottom of the chain band rises above the Fermi level. We note that when the asymptotic behaviors Eqs. (23) and (27) are valid we merge smoothly into a regime where a simple standard BCS formula with the density of states calculated at $E_F$ can be used for the various contributions in Eq. (9). This was naturally expected.

From the various contributions which we have obtained it is clear that the effect of departing from the ideal situation is not at all dramatic, as one might fear, but rather progressive. Naturally the energy scale for all the effects is $2k_B T_c$ but on this scale they are rather weak, whereas one could think that they are strong. To be specific we consider various situations to illustrate our point. We compare again the critical temperature with ($T_c^0$) and without ($T_c^1$) the chain band always starting from $T_c^0 = 95$ K and looking for $T_c^1$.

Let us first keep the Fermi level at the middle of the plane band $E_F = 0$. We have to solve Eq. (17) where the right-hand side is replaced by $F(Y_F)$. If we raise the bottom of the chain band $T_c^0/T_c^1$ decreases rapidly. For the given value of $T_c^0 = 95$ K, we reach $T_c^1 = 40$ K for $\varepsilon_0 \approx 0.022$ eV (and $T_c^1 = 95$ K for $\varepsilon = 0.05$ eV since the cutoff $\omega_0 = 0.05$ eV). If we lower the bottom of the chain band, $T_c^0/T_c^1$ first rises and for $\varepsilon_0 = -0.016$ eV we obtain $T_c^1 = 17$ K. Then $T_c^0/T_c^1$ decreases, but $T_c^1 = 40$ K is reached only for $\varepsilon_0 = -0.11$ eV. Therefore there is a fairly wide range (0.13 eV) compared to $2k_B T_c^0$ (0.016 eV) where $T_c^0/T_c^1$ is larger than the experimental value.

Then we take the Fermi level away from the middle of the plane band. This will actually increase $T_c^0/T_c^1$ since the effective density of states of the plane band is decreased compared to the chain band one. But obviously this situation is much less favorable for high $T_c$. One can study with Eqs. (18)–(23) the effect on $T_c$ of shifting away the Fermi level, but this is not our purpose in this paper (one would need to know the interaction $V$ in order to make such a study effective). Let us just indicate that, as expected, an important decrease in $T_c$ is obtained when $E_F$ shifts by a few $2k_B T_c^0$.

Coming back to $T_c^0/T_c^1$ when the asymptotic behavior Eq. (23) can be used, we obtain a simple BCS result for the plane band and we have to merely solve

$$\ln(T_c^0/T_c^1) = v_2 b_0^{1/2} F(Y_F^0) / 2 \ln(t_1 e^{v_1}/|E_F|) , \tag{30}$$

where $Y_F^0$ is $Y_F$ with $T_c = T_c^0$. If we take the bottom of the chain band at the Fermi level ($Y_F^0 = 0$) we have

$$\ln(T_c^0/T_c^1) = 11.6 / \ln(t_1 e^{v_1}/|E_F|) . \tag{31}$$

For $|E_F| = 0.05$ eV, $T_c^1 = 9.5$ K becomes naturally smaller than what we found for $E_F = 0$. If we raise the bottom of the chain band $T_c^1 = 40$ K is reached for $\varepsilon_0 - E_F = 0.03$ eV. If we lower it below the Fermi level, we reach soon a simple BCS regime and

$$\ln(T_c^0/T_c^1) = v_2 \left( \frac{t_1}{E_F - \varepsilon_0} \right)^{1/2} \frac{(\alpha + \ln \Omega_0)}{\ln(t_1 e^{v_1}/|E_F|)} . \tag{32}$$

For $|E_F| = 0.05$ eV, we obtain $T_c^1 = 40$ K when $E_F - \varepsilon_0 = 0.24$ eV (this result is not very sensitive to our choice of $E_F$). Therefore for $|E_F| = 0.05$ eV we obtain that $T_c^0/T_c^1$ is larger than experiment when the bottom of the chain band varies in an energy range of width 0.27 eV.

From the above discussion it is clear that we obtain a $T_c^0/T_c^1$ much larger than experiment for a fairly large range of the parameters $\varepsilon_0$ and $E_F$ of typical size 0.1 eV. Therefore it one takes into account the broadening effects of interchain and interplane coupling, the experimental result can still be explained by our model provided that this broadening is of order 0.1 eV (which is reasonable as we have seen). Indeed broadening effects amount to shift around the energy of the eigenstates. Therefore the result is qualitatively similar to a shift in the Fermi energy. Similarly, scattering gives a broadening over an energy range $\hbar/\tau$. The relaxation time can be estimated from the plasma frequency and the conductivity by $\tau = 4\pi\sigma/\omega_p^2$. With $\omega_p = 2$–$3$ eV from the infrared data and $1/\sigma \approx 10^2$ $\mu\Omega$ cm, one gets $\hbar/\tau \approx 0.1$ eV and broadening due to scattering is not strong enough to modify our conclusion.

Let us make a few concluding remarks. One can consider our calculation Eq. (9) as obtaining an effective density of states. It is important to realize that these effective density of states are obtained by integrating the actual density of states over a rather large energy interval of order $4k_BT_c$. Moreover the density of states beyond that range contribute in a very important way, typically its weight is half of the integral in Eq. (9). Therefore we believe that if the results from band-structure calculations are properly integrated, they will agree with our conclusion that the increase in effective density of states is enough to explain the increase in $T_c$ in going from $La_{2-x}Sr_xCuO_4$ to $YBa_2Cu_3O_7$. Moreover this averaging process could bring the final results of the various band-structure calculations in much better agreement than they are presently.

Our use of the weak-coupling theory can certainly be criticized. Indeed taking as an average phonon frequency $\langle\omega\rangle$ our cutoff $\omega_0 = 0.05$ eV one obtains[25] for $T_c = 95$ K an effective coupling constant $\lambda = 1.6$ (for $\mu^* = 0$) which is a typical value[26] suggested for the high-$T_c$ compounds. A higher $\lambda$ is obtained for a lower $\langle\omega\rangle$. However, it is known[25] that, at fixed $\langle\omega\rangle$, the critical temperature $T_c$ grows as $\lambda^{1/2}$ for large $\lambda$ and does not saturate as does the McMillan formula. The naive weak-coupling theory gives[25] $T_c = \omega_D\lambda/2$ for large $\lambda$. Therefore it is reasonable to believe that our results for the increase in critical temperature, that is the ratio $T_c^0/T_c^1$, would not be too much reduced by a proper analysis with strong-coupling theory.

Another point is that the densities of states which we use are reasonable and in agreement with the experimental estimates for the specific heat. Indeed it must be realized that in order to obtain the specific heat at temperature $T$, one averages the density of states over a range $4k_BT$ around the Fermi level. Therefore in our ideal situation, we can take the density of states at $2k_BT$ from $E_F$ and use it as an effective density of states in the Sommerfeld constant. Actually,[27]

$$C(T) = \frac{1}{4k_BT^2} \int d\varepsilon \frac{\varepsilon^2 N(\varepsilon)}{ch^2(\varepsilon/2k_BT)} , \tag{33}$$

with $N(\varepsilon) = 2N_1(\varepsilon) + N_2(\varepsilon)$ given by Eqs. (5) and (6), leads exactly to[28]

$$C_1(T) = \frac{\pi^2}{3} k_B^2 T \left( \frac{2N}{2\pi^2 t_1} \right) \left[ \ln \left( \frac{16 t_1}{2k_BT} \right) - 0.35 \right] , \tag{34}$$

$$C_2(T) = \frac{\pi^2}{3} k_B^2 T \left( \frac{2N}{2\pi t_2} \right) \frac{1}{2} \left( \frac{t_2}{2k_BT} \right)^{1/2} \times 0.87 , \tag{35}$$

with $C(T) = 2C_1(T) + C_2(T)$. Taking merely the density of states at $2k_BT$ is therefore a good approximation since it amounts to neglect of the additional term 0.35 in Eq. (34) and the factor 0.87 in Eq. (35) [the factor $\frac{1}{2}$ in Eq. (35) comes because we have $N_2(\varepsilon) \neq 0$ for $\varepsilon > 0$ only whereas the density of states $N_1(\varepsilon)$ is symmetrical]. Now for $T = 95$ K, Eqs. (34) and (35) correspond to an effective density of states of 3.90 states/cell/eV for the two spin directions together, equivalent to an effective Sommerfeld constant of 9.2 mJ/mol K$^2$ which is typically half of the current experimental estimates.[29] We note also that in order to have $T_c = 95$ K in Eq. (15) we need $I = 48$ which gives the reasonable value 0.20 eV × cell for the parameter $NV$.

Finally we note that the isotopic effect is easily obtained by differentiating Eq. (7). This leads in general to

$$\frac{d\omega_0}{dT_c} \frac{T_c}{\omega_0} = \frac{1}{N(\omega_0) + N(-\omega_0)} \int_{-\infty}^{+\infty} dy \frac{N(2k_BT_cy)}{ch^2 y} . \tag{36}$$

Roughly speaking $\omega_0 dT_c/T_c d\omega_0$ is the ratio of the density of states at $\omega_0$ to the density of states at $2k_BT_c$, and therefore it is reduced with respect to the standard BCS value of 1. In the present case we obtain

$$\frac{dT_c}{d\omega_0} \frac{\omega_0}{T_c} = \frac{\ln(r_0 e^{v_1}) + v_2 r_0^{1/2}/2}{\ln(b_0 e^{\alpha + v_1}) + v_2 b_0^{1/2} L(0)/4} = 0.49 . \tag{37}$$

This trend toward reduction agrees with the experimental results.[30] However, it is known that Coulomb repulsion plays an important role in the isotopic effect, and it is therefore not surprising that we do not obtain the nearly zero isotopic effect found experimentally in $YBa_2Cu_3O_7$. Since the isotopic effect is notoriously very difficult to calculate with rather uncertain results we cannot draw any further conclusion.

In conclusion we find that the increase in $T_c$ from $La_{2-x}Sr_xCuO_4$ to $YBa_2Cu_3O_7$ or the decrease in $T_c$ due to oxygen depletion in $YBa_2Cu_3O_7$ can be easily explained by a change in the effective density of states, obtained in a natural way from a simple band-structure calculation. It is therefore reasonable to believe, or at least impossible to exclude, that density-of-states effects are partly responsible for the high values of the critical temperature itself, together with the known high value of the phonon energies in these compounds.

## ACKNOWLEDGMENT

We are grateful to J. Bok and P. Monod for discussions.

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[1] J. G. Bednorz and K. A. Müller, Z. Phys. B **64**, 189 (1986); C. W. Chu, P. H. Hor, R. L. Meng, L. Gao, Z. J. Huang, and Y. Q. Wang, Phys. Rev. Lett. **58**, 405 (1987).

[2] T. M. Rice, Z. Phys. B **67**, 141 (1987).

[3] J. Labbe, S. Barisic, and J. Friedel, Phys. Rev. Lett. **19**, 1039 (1967).

[4] J. E. Hirsch and D. J. Scalapino, Phys. Rev. Lett. **56**, 2732 (1986).

[5] J. Labbe and J. Bok, Europhys. Lett. **3**, 1225 (1987); J. D. Jorgensen, H. B. Schüttler, D. G. Hinks, D. W. Capone II, K. Zhang, M. B. Brosky, and D. J. Scalapino, Phys. Rev. Lett. **58**, 1035 (1987); H. Fukuyama and Y. Hasegawa, J. Phys. Soc. Jpn. (to be published).

[6] J. J. Caponi, C. Chaillout, A. W. Hewat, P. Lejay, M. Marezio, N. Nguyen, B. Raveau, J. L. Soubeyroux, J. L. Tholence, and R. Tournier, Europhys. Lett. **3**, 1301 (1987); W. K. Kwok, G. W. Crabtree, A. Umezawa, B. W. Veal, J. D. Jorgensen, S. K. Malik, L. J. Nowicki, A. P. Paulikas, and L. Nunez (to be published).

[7] S. Barisic, I. Batistic, and J. Friedel, Europhys. Lett. **3**, 1231 (1987).

[8] L. F. Mattheiss, Phys. Rev. Lett. **58**, 1028 (1987).

[9] J. Yu, A. J. Freeman, and J. H. Xu, Phys. Rev. Lett. **58**, 1035 (1987).

[10] S. Massida, J. Yu, A. J. Freeman, and D. D. Koelling, Phys. Lett. **122**, 198 (1987).

[11] J. Yu, S. Massida, A. J. Freeman, and D. D. Koelling, Phys. Lett. **122**, 203 (1987).

[12] M. H. Whangbo, M. Evain, M. A. Beno, and J. M. Williams, Inorg. Chem. **26**, 1831 (1987); **26**, 1832 (1987).

[13] L. F. Mattheis and D. R. Hamann, Solid State Commun. **63**, 395 (1987).

[14] F. Herman, R. V. Kasowski, and W. Y. Hsu, Phys. Rev. B (to be published).

[15] Such a near ideal situation has already been discussed by J. Friedel, C.R. Acad. Sci. Paris **305**, 543 (1987).

[16] Most band calculations also find a narrow Cu$d_{xz}$-O$p_z$-O$p_x$ band due to the chains which lies near $E_F$. Since its physical meaning is unclear, we do not retain it in our model (we would also expect a lower energy for this band). Needless to say, taking its density of states into account would help in explaining high $T_c$ for $YBa_2Cu_3O_7$.

[17] P. Monod, M. Ribault, F. D'yvoire, J. Jegoudez, G. Collin, and A. Revcolevschi, J. Phys. **48**, 1369 (1987).

[18] D. C. Johnston, A. J. Jocobson, J. M. Newsam, M. T. Lewandowski, and D. P. Goshorn, in *Chemistry of High Temperature Superconductors,* ACS Symposium Series, No. 351, edited by D. L. Nelson, M. S. Whittingham, and T. F. George (American Chemical Society, Washington, DC, 1987).

[19] J. Labbe and E. C. van Reuth, Phys. Rev. Lett. **24**, 1232 (1970).

[20] Here we neglect the splitting of the Van Hove singularity due to the small orthorhombic distortion in $La_{2-x}Sr_xCuO_4$. Since the density of states has to be averaged over typically $4k_BT_c$ and because of impurity smearing, this approximation is not likely to be very important. It could be studied quantitatively with Eq. (18) to Eq. (23). The effect of the doping $x$ is not very important. For example, if we take the doping $x=0.15$ and assume a fixed Fermi level, then we have for $YBa_2Cu_3O_7$ 2.3 holes in the plane bands and 1.7 holes in the chain band. This corresponds to $\varepsilon_0=-0.12$ eV which falls in the range of parameters that we have studied.

[21] I. S. Gradshteyn and I. W. Ryzhik, *Tables of Integrals, Series and Products* (Academic, New York, 1965).

[22] Equation (5) is actually a good approximation of Eq. (4) over the whole range of energy: the integrated density of states is $4N\ln(4e)/\pi^2=0.967N$ instead of the exact result $N$ and $N(0)=N\ln 4/2\pi^2 t=0.070N/t$ instead of $N/4\pi t=0.079N/t$.

[23] P. Monod (private communication).

[24] Actually if for $YBa_2Cu_3O_7$ the Fermi level is located above the bottom of the chain band, we will have a shift of the Fermi level when the chain band will be destroyed by oxygen removal because we have to transfer the electrons from the chain band to the plane band. However, this shift will either increase or decrease $T_c^0/T_c^1$ depending on the location of the Fermi level with respect to the maximum of the density of states in the plane band. Therefore we ignore this shift for simplicity, since our aim is to find the typical energy scale for $\varepsilon_0$ on which $T_c^0/T_c^1$ is in agreement with the experimental results.

[25] P. B. Allen and R. C. Dynes, Phys. Rev. B **12**, 905 (1975).

[26] W. Weber, Phys. Rev. Lett. **58**, 1371 (1987); B. Ashauer, W. Lee, and J. Rammer, Z. Phys. B **67**, 15 (1987).

[27] This neglects the effect of the temperature dependence of the chemical potential. But this is a small effect because it comes only from the chain band since the density of states for the plane band is symmetrical. The correction on our numerical result is of order 10%.

[28] J. Bok and J. Labbe, C.R. Acad. Sci. Paris **305**, 555 (1987).

[29] A. Junod *et al.*, Europhys. Lett. **4**, 247 (1987).

[30] K. J. Leary, H-C. zur Loye, S. W. Keller, T. A. Faltens, W. K. Ham, J. N. Michaels, and A. M. Stacy, Phys. Rev. Lett. **59**, 1236 (1987).