Dependence of T_c on the physical parameters in single-layered copper oxides

T. Hocquet

Laboratoire de Physique de la Matière Condensée de l'École Normale Supérieure, 24 rue Lhomond, 75231 Paris Cedex 05, France

J.-P. Jardin

Laboratoire des Propriétés Mécaniques et Thermodynamiques des Matériaux (CNRS), Université Paris Nord, 93430 Villetaneuse, France

P. Germain and J. Labbé
Laboratoire de Physique de la Matière Condensée de l'École Normale Supérieure, 24 rue Lhomond, 75231 Paris Cedex 05, France

(Received 21 February 199S)

From the superconducting gap equation that we have derived in a previous paper, we calculate the critical superconducting temperature T_c of an itinerant electron gas in a CuO₂ plane as a function of the occupancy of the d-p subband. This involves applying the Bogoliubov-Valatin mean-field approximation to a Hamiltonian including both the Hubbard intra-atomic repulsion U and an effective coupling constant V between electrons; the latter is assumed to be mediated by the electron-phonon interaction. As the Fermi level is shifted from the logarithmic Van Hove singularity in the electronic density of states, the calculated value of T_c decreases much more slowly than the antiferromagnetic order parameter that we had previously calculated in the same model. This explains the existence of the superconducting phase largely outside of the domain of stability of the antiferromagnetic phase, when the compound is doped. Furthermore, we calculate the isotope effect, which exhibits a large variation with the doping x , showing a sharp peak for some critical value of x , in agreement with several independent experimental data. A very large influence of the Coulomb parameter U is found on the isotope effect which, in some cases, could vanish, or even be negative.

I. INTRODUCTION

We use the tight-binding model which had been initially proposed by Labbé and Bok¹ for calculating the critical superconducting temperature T_c of an itinerant electron gas in a $CuO₂$ plane. This model is supported by very recent experimental photoemission data^{2,3} which clearly show, not only the existence of a Fermi energy, but also the existence of a bidimensional saddle point in the energy spectrum, indicating the itinerant nature of the electron gas and its bidimensional character. In their paper, Labbe and Bok had predicted a reduction of the isotope effect when the Fermi energy nearly coincides with the logarithmic Van Hove singularity in the electronic density of states. More recently, Tsuei et $al.$ ⁴ used this model for calculating the variations of T_c when the Fermi energy is shifted from the singularity. As an interesting result, they found that the isotope effect strongly depends on the Fermi-level position. But in their calculation they did not take into account the repulsive part of the Hamiltonian, which is known to be essential to explain other properties of such systems, as for instance the existence of an antiferromagnetic phase. In the present paper, we demonstrate that if we also include the intraatomic Coulomb repulsion between itinerant electrons, the variations of T_c and of the isotope effect, when the occupancy of the band is varied, are strongly modified. In previous papers,^{5,6} we discussed the stability of the antiferromagnetic phase in a single $CuO₂$ plane, as for instance in $La_{2-x}Sr_xCuO_4$, and we calculated the superconducting gap at the absolute zero versus the doping ratio x . We explained why the antiferromagnetic phase was confined to small values of x only. On the contrary, we found a slow variation of the calculated superconducting gap versus x . The equation for calculating the superconducting gap at any temperature T is reintroduced in Sec. II, the variations of T_c versus x are calculated in Sec. III, and the predictions of the model concerning the isotope effect are extensively discussed in Sec. IV, with a comparison to the existing experimental results.

II. EQUATION FOR THE SUPERCONDUCTING GAP

In our model, the interaction between two electrons with wave vectors k and k' is made of two contributions: the first is the effective attractive interaction $V_{kk'}$ and the second is the Coulomb repulsion U between two electrons on the same atomic site with opposite spins, according to the Hubbard model. Solving the total Hamiltonian containing these two contributions in the Bogoliubov and Valatin mean-field approximation,⁶ we got the following equation for the superconductivity gap Δ_k .

$$
\Delta_{\mathbf{k}} = -\sum_{\mathbf{k}'} \left[V_{\mathbf{k}\mathbf{k}'} + \frac{U}{N} \right] \frac{\Delta_{\mathbf{k}'}}{2\mathscr{E}_{\mathbf{k}'}} \tanh\left(\frac{\mathscr{E}_{\mathbf{k}'}}{2k_B T}\right), \quad (1)
$$

where $\mathscr{E}_k = \sqrt{\epsilon_k^2 + \Delta_k^2}$ is the energy of an elementary excitation, $\varepsilon_k = E_k + \frac{Un}{2} - \mu$ is the Hartree-Fock oneparticle energy referred to the chemical potential μ , E_k

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the bare particle energy in the band, $n = 1-x$ the averaged number of itinerant electrons per copper atom, T the temperature, and N the number of unit cells.

We assumed that $V_{kk'}$ has a nonvanishing value

 $-V/N$, with $V>0$, only if both $|\epsilon_{\mathbf{k}}|$ and $|\epsilon_{\mathbf{k'}}|$ are smaller than some cutoff energy $\hbar\omega_0$. Then it results from Eq. (1) that the energy gap Δ_k has two distinct constant values Δ_1 and Δ_2 according to the sign of $|\epsilon_{\mathbf{k}}| - \hbar \omega_0$, with

$$
\Delta_{\mathbf{k}} = \begin{cases}\n\Delta_1 = V \Delta_1 F(T, \Delta_1) - U(\Delta_1 F(T, \Delta_1) + \Delta_2 G(T, \Delta_2)) & \text{if } |\varepsilon_{\mathbf{k}}| \leq \hbar \omega_0, \\
\Delta_2 = -U(\Delta_1 F(T, \Delta_1) + \Delta_2 G(T, \Delta_2)) & \text{if } |\varepsilon_{\mathbf{k}}| > \hbar \omega_0,\n\end{cases}
$$
\n(2)

with

$$
F(T, \Delta_1) = \frac{1}{N} \sum_{|\varepsilon_k| \leq \hbar \omega_0} \frac{1}{2\mathcal{E}_k} \tanh\left(\frac{\mathcal{E}_k}{2k_B T}\right),
$$

\n
$$
G(T, \Delta_2) = \frac{1}{N} \sum_{|\varepsilon_k| > \hbar \omega_0} \frac{1}{2\mathcal{E}_k} \tanh\left(\frac{\mathcal{E}_k}{2k_B T}\right).
$$
\n(3)

We applied these equations to a CuO₂ plane, with our model density of states, for $|E| \leq 4t$,

$$
\mathcal{D}(E) = \frac{1}{2\pi^2 t} \ln \frac{16t}{|E|} \tag{4}
$$

in which $E=\epsilon-Un/2+\mu$, and t is the effective transfer integral between neighboring copper atoms.⁷

From these equations, we had calculated the gaps $\Delta_1(0)$ and $\Delta_2(0)$ for $T = 0$ K, the striking feature of our results being that $\Delta_1(0)$ and $\Delta_2(0)$ decrease slowly when the doping ratio x increases.⁶

III. CALCULATED VARIATIONS OF T_c VERSUS x

Equations (2) always have a trivial solution $\Delta_1 = \Delta_2 = 0$ and, in the superconducting state, another nonvanishing one, which leads to a more stable state. At the critical superconducting temperature T_c , this second solution itself vanishes. This leads to the following implicit equation to determine T_c .

$$
(UG_0(T_c)+1)(VF_0(T_c)-1)=UF_0(T_c), \qquad (5)
$$

with $F_0(T_c) = F(T_c, 0)$ and $G_0(T_c) = G(T_c, 0)$. In the case of a CuO₂ plane, our model density of states (4) gives

$$
F_0(T_c) = \frac{1}{4\pi^2 t} \int_{-\hbar\omega_0}^{\hbar\omega_0} \ln \left| \frac{16t}{\epsilon - \delta} \right| \tanh \frac{\epsilon}{2k_B T_c} \frac{d\epsilon}{\epsilon}
$$

\n
$$
G_0(T_c) = \frac{1}{4\pi^2 t} \left\{ \int_{-4t + \delta}^{-\hbar\omega_0} \ln \left| \frac{16t}{\epsilon - \delta} \right| \tanh \frac{\epsilon}{2k_B T_c} \frac{d\epsilon}{\epsilon} + \int_{\hbar\omega_0}^{4t + \delta} \ln \left| \frac{16t}{\epsilon - \delta} \right| \tanh \frac{\epsilon}{2k_B T_c} \frac{d\epsilon}{\epsilon} \right|,
$$
\n(6)

where $\delta = Un/2 - \mu$ is the opposite of the shift of the chemical potential with respect to the logarithmic singularity of $\mathcal{D}(E)$. At T_c , this shift is related to the doping ratio x through the Fermi-Dirac distribution, valid in the normal state:

$$
1 - x = \frac{1}{2\pi^2 t} \int_{-4t + \delta}^{4t + \delta} \ln \left| \frac{16t}{\epsilon - \delta} \right| \frac{d\epsilon}{1 + e^{\epsilon/k_B T_c}} \ . \tag{7}
$$

The procedure to get T_c versus x is, for each chosen value of δ , to calculate T_c from Eq. (5), and then x from Eq. (7). Figure ¹ shows our numerical results for different values of U. Figure 2 shows the calculated variation of x versus δ : we see that this variation is nearly independent of the value of T_c , and thus of U, ω_0 and V, except for very small doping x .

The value of T_c is very sensitive to U: T_c is reduced by a factor \sim 5 when going from $U = 0$ to $U = 2t$. We do not know accurately the numerical values of the parameters. But in our model, only the dimensionless parameters U/t , V/t , and $\hbar \omega_0/t$ are relevant to determine the reduced value $k_B T_c/t$ as a function of x. In fact, the calculated values of T_c for $U=0$ are very large: for $V=2t$, $\hbar \omega_0 = 0.2t$, one finds $k_B T_c = 0.022t$ for $x = 0.15$, which gives $T_c \approx 130$ K if, for instance, $t = 0.5$ eV, which is too large as compared to the experimental values. On the contrary, with $U = t$ and the same values of the other parameters, we get more realistic values of T_c , $T_c \simeq 40$ K for $x = 0.15$, comparable to the experimental values observed in $\text{La}_{2-x} \text{Sr}_x \text{CuO}_4$. Obviously, these results do not take into account the fact that, in a single $CuO₂$ plane, the antiferromagnetic phase is more stable for x smaller than a small critical value, as shown in one of our previous papers,⁵ forbidding the superconducting phase to exist in this narrow range. But in the present paper, we focus our attention on the predictions of the model only for superconductivity, as if the antiferromagnetic phase would not exist.

FIG. 1. Reduced superconductor critical temperature $k_B T_c/t$ versus the doping ratio x, exactly calculated (full line) and by the approximate formula (9) (dashed line), for $V/t = 2$, $\hbar \omega_0/t = 0.2$, and for $U=0$ (curve a), $U/t=1$ (curve b), and $U/t = 2$ (curve c).

As $G_0(T_c)$ depends much less on δ than $F_0(T_c)$ does,⁶ it can be replaced by the simplified version it would have at $x = 0$. Then, by integrating by parts the second Eq. (6), and assuming $k_B T_c \ll \hbar \omega_0$, one gets

$$
G_0(T_c) \simeq \frac{1}{4\pi^2 t} \ln \frac{4t}{\hbar \omega_0} \ln \frac{64t}{\hbar \omega_0} \ . \tag{8}
$$

As in this case $G_0(T_c)$ in fact does not depend on T_c , Eq. (S) reduces to

$$
(V - U^*)F_0(T_c) \simeq 1 , \t\t(9)
$$

FIG. 2. The doping ratio x versus the opposite of the Fermilevel shift δ , for $\hbar \omega_0/t = 0.2$, $U = 0$, and $V/t = 2$, at $T = T_c$ (full line) and at $T = 0$ K (dashed line).

where the effective Coulomb repulsive parameter

$$
U^* = \frac{U}{1 + (U/4\pi^2 t) \ln(4t/\hbar\omega_0) \ln(64t/\hbar\omega_0)}
$$
(10)

is the same as previously found when calculating the gap. 6 The simplified equation (9) leads to a very good approximation for the calculated values of T_c versus x, as compared to the exact equation (S) (see dashed lines in Fig. 1).

The effect of U on T_c can be explicitly analyzed for $x = 0$. In this case, when integrating by parts the integrals in the first expression (6) with $k_B T_c \ll \hbar \omega_0$, one gets

$$
F_0(T_c) \simeq \frac{1}{2\pi^2 t} \left\{ \frac{1}{2} \left[\ln \frac{\hbar \omega_0}{2k_B T_c} \right]^2 + \left[a + \ln \frac{16t}{\hbar \omega_0} \right] \ln \frac{\hbar \omega_0}{2k_B T_c} + b + a \ln \frac{16t}{\hbar \omega_0} \right\},
$$
\n(11)

with $a = C - \ln(\pi/4) \approx 0.82$, where C is the Euler constant, and $b \approx 0.99$. Then, a straightforward calculation leads to the explicit expression

$$
k_B T_c \simeq \frac{32t}{\pi} e^{C} \exp\left\{-\left[\left[\ln \frac{16t}{\hbar \omega_0}\right]^2 + \frac{4\pi^2 t}{V - U^*} - 1.32\right]^{1/2}\right\},\tag{12}
$$

which explicitly shows the strong effect of U on T_c at $x = 0$.

The exponential factors in the expression (10) of T_c and in the expression (34) of our previous paper for $\Delta_1(0)$ at $T = 0$ K are almost identical. Thus the ratio $2\Delta_1(0)/k_B T_c$ is essentially equal to the ratio of the two prefactors in front of the exponentials in these expressions, and in fact has the standard BCS value $2\pi e^{-C} \approx 3.53$. Numerical calculation shows that the value of this ratio remains practically constant when the doping ratio increases.

It is clear that our model is not able to explain a ratio $2\Delta_1(0)/k_B T_c$ which would be much larger than the usual BCS value. A large value, if it was experimentally confirmed, could be explained either by the Eliashberg strong-coupling theory, 8 or by an effect of large bidimensional fluctuations which would reduce T_c but not $\Delta_1(0)$, or also by a large anisotropy of the attractive coupling $\mathop{\rm constant}\nolimits V_{\bf kk'}$

IV, INFLUENCE OF THE CUTOFF ENERGY $\hbar\omega_0$

In our model, the effect of $\hbar \omega_0$ on T_c can be calculated. It must be emphasized that our theory, which is of BCS type, does not necessarily assume that the attractive part of the interaction between two electrons is induced by phonons: in principle it could be induced as well by some other boson field. But in the case of an electron-phonon mechanism, the calculation of the effect of $\hbar \omega_0$ leads to the discussion of the isotope efFect.

The dependence of T_c on ω_0 , for a given value of the doping ratio x , can be calculated by differentiating Eq. (5) with respect to T_c and ω_0 in which δ depends on T_c through Eq. (7). Qne finds

$$
\frac{dT_c}{d\omega_0} = \frac{(VF_0 - 1)^2 - 1}{\partial F_0 / \partial T_c + (VF_0 - 1)^2 (\partial G_0 / \partial T_c)} \frac{\partial F_0}{\partial \omega_0}, \quad (13)
$$

where we took advantage of the equality $\partial G_0/\partial \omega_0 = -\partial F_0/\partial \omega_0$. When deriving Eq. (13), we started from the exact equation (5), and not from its simplified version (9), because we have now to calculate very small variations of T_c versus ω_0 , and not the absolute value of T_c itself. The partial derivatives of F_0 and G_0 with respect to ω_0 and T_c , for a constant value of x, can be calculated from Eqs. (6) and (7). The calculation is greatly simplified by doing approximations which use the fact that the subband width $8t$ is much larger than the cutoff energy $\hbar \omega_0$, which itself is assumed to be much larger than $k_B T_c$. The resulting explicit expression of the coefficient $\gamma = \partial \ln T_c / \partial \ln \omega_0$, which is given in Appendix A, makes possible numerical calculations of the value of γ as function of the physical parameters of the model. The most striking feature of our results is that γ does not have the usual constant value, equal to unity, which would be predicted by the simplest version of the BCS model, but on the contrary it exhibits a large variation versus the doping ratio x , and very critically depends on the respective values of the repulsive Coulomb parameter U and the effective attractive coupling constant V . According to the choice of the parameters, γ can be either much smaller or much larger than unity, it can vanish, even with an electron-phonon mechanism, or also be negative. Figure 3 shows the calculated variations of γ versus x for different choices of the parameter U .

For $x = 0$ [and thus $u_0 = 0$ in Eq. (A2)], the expression for γ reduces to

$$
\gamma = \frac{d \ln T_c}{d \ln \omega_0} \simeq \frac{V(V - 2U^*)}{(V - U^*)^2} \frac{\ln(16t / \hbar \omega_0)}{0.82 + \ln(8t / k_B T_c)},
$$
\n(14)

where we took advantage of the simplified equation (9), with U^* given by Eq. (10). The expression (14) shows that when the Fermi energy exactly coincides with the logarithmic singularity in the density of states, the more drastic modification of the isotope efFect comes from the presence of the effective repulsive potential U^* . For instance, for $2U^*$ > V, the expression (14) is negative, and thus the isotope effect is reversed. On the contrary, for U^* =0, the expression (14) is always positive, and for realistic values of the other parameters, the reduction of the isosope effect by the factor $\ln(16t/\hbar\omega_0)/(0.82)$

FIG. 3. Logarithmic derivative $\gamma = \partial \ln T_c / \partial \ln \omega_0$ versus the doping ratio x, calculated for $V/t = 2$, $\hbar \omega_0/t = 0.2$, and for $U=0$ (curve a), $U/t=1$ (curve b), $U/t=1.88$ (curve c), and $U/t = 2$ (curve **d**).

 $+\ln(8t/k_B T_c)$, which is due to the decrease of the electronic density of states when going from μ to $\mu \pm \hbar \omega_0$, cannot be very large, because the singularity has a logarithmic behavior only.¹⁰

The very sharp peak found in the calculated variations of γ for larger values of x in Fig. 3, which exists whatever the value of U is, is explained by the fact that, when the logarithmic singularity in the electronic density of states is very close to one of the two limits $\mu \pm \hbar \omega_0$ of the energy range inside which the attractive interaction exists, a small variation of ω_0 produces a large change in the value of T_c . We show in Appendix B that when δ is very close to $\hbar\omega_0$, the expression for γ exhibits a logarithmic singularity, and is approximately given by B that when δ is very close
hibits a logarithmic singu-
en by
 $\frac{\pi\omega_0 + \ln(16t/|\pi\omega_0-\delta|)}{2\ln(16t/|\pi\omega_0|)},$

$$
\gamma \simeq [1 - (VF_0(T_c) - 1)^2] \frac{\ln(8t/\hbar\omega_0) + \ln(16t/\hbar\omega_0 - \delta|)}{2\ln(16t/\hbar\omega_0)},
$$
\n(15)

where we must keep the exact expression of $F_0(T_c)$, which depends on U through Eq. (5), because it is very sensitive to the value of x for large doping. We see from Eqs. (A1) and (A2), that γ vanishes at a critical value of x, if it exists, for which $VF_0=2$, leading to an infinite value of the denominator D of the expression $(A1)$.

The quantitative comparison of the above theoretical predictions to existing experimental results $11-14$ is difficult because the relation between ω_0 and the masses of the different ions in such compounds is not clearly established. Nevertheless, it has been recently observed that, in $La_{2-x}Sr_xCuO_4$, the substitution of ⁶⁵Cu to ⁶³Cu leads to an isotope effect of the same magnitude as the substitution of ^{18}O to ^{16}O , showing that large parts of the phonon spectrum inhuence the superconducting transition temperature.¹⁴

Almost all the experimental data in $\text{La}_{2-x} A_x \text{CuO}_4$ $(A = Sr, Ba)$ show a very large influence of the doping on the isotope effect: a sharp peak is observed in the variations of the latter with respect to the doping x , in agreement with our calculations (Fig. 3). As we do not know the precise relation between the ion masses and the phonon frequencies, we cannot hope to fit in detail the experimental data with our continuous calculated curves. However, the position of our peak in Fig. 3 coincides with the experimental one $(x \approx 0.12)$ if we assume $\hbar\omega_0 \approx 0.215t$, which is a very reasonable value for the maximum phonon frequencies, with, for instance, $t = 1$ eV. It can be noticed that, in our model, the position of the peak depends only very little on U and V . This peak has not been observed in $YBa₂Cu₃O_{6+y}$. But in this last compound, there is not a continuous shift of the Fermi level when the oxygen content is gradually modified: on the contrary, for a given value of y (roughly equal to 0.4), there is a sudden transfer of a finite number of electrons from the $CuO₂$ planes to the CuO linear chains appearing in the structure at this composition. Some authors had attributed the existence of this peak in $\text{La}_{2-x} \text{Sr}_x \text{CuO}_4$ to a structural phase transition which would be induced by doping. But recent high pressure studies¹⁵ and the comparison of the effects of copper and oxygen isotope substitution¹⁴ show that the observed peak can hardly be attributed to structural instabilities.

A physical conclusion of our theoretical study is that a very small, or even vanishing, isotope effect is not in contradiction with an electron-phonon mechanism for the superconductivity in these compounds.

APPENDIX A

By calculating the partial derivatives of F_0 and G_0 from their expressions (6), and assuming $k_B T_c$ $\ll \hbar\omega_0 \ll 8t$, the expression (11) can be transformed to

$$
\gamma \simeq \frac{d \ln T_c}{d \ln \omega_0} = \frac{1}{D} \ln \frac{(16t)^2}{|(\hbar \omega_0)^2 - \delta^2|} , \qquad (A1)
$$

where

$$
D = \frac{1}{1 - (VF_0 - 1)^2} \left\{ 2 \ln \frac{8t}{k_B T_c} - K(u_0, +\infty) + \frac{d \ln \delta}{d \ln T_c} [K(u_0, +\infty) - K(0, +\infty)] \right\} + K(u_0, +\infty) - K(u_0, u_1)
$$

$$
- \frac{d \ln \delta}{d \ln T_c} \left[K(u_0, +\infty) - K(0, +\infty) + K(u_0, u_1) + K(0, u_1) + \ln \left| 1 - \left(\frac{\delta}{\hbar \omega_0} \right)^2 \right| \right],
$$
(A2)

with
$$
u_0 = \delta / 2k_B T_c
$$
, $u_1 = \hbar \omega_0 / 2k_B T_c$, and

$$
K(u_0, u_1) = \int_{-u_1}^{+u_1} \ln|u - u_0| \frac{du}{\ch^2 u}
$$

Then, by taking the derivative of Eq. (7) with respect to T_c , for a given value of x, with the same approximations as above, one gets

$$
\frac{d \ln \delta}{d \ln T_c} = \frac{k_B T_c}{\delta} \frac{\Lambda(u_0)}{2 \ln \frac{8t}{k_B T_c} - K(u_0, +\infty)},
$$
 (A3)

with

$$
\Lambda(u_0) = -\int_{-\infty}^{+\infty} \ln|u - u_0| \frac{u \, du}{\mathrm{ch}^2 u} \; .
$$

The three equations (A1), (A2), and (A3) determine γ and make possible numerical calculations. One can use the approximate value $K(0, +\infty) \approx -1.64$, and also the

asymptotic behaviors $K(u_0, u_1) \approx 2 \ln u_0$ and $\Lambda(u_0)$ $\simeq \pi^2/6u_0$, valid for $u_0 \gg 1$ and $u_1 \gg 1$.

APPENDIX B

If δ would be exactly equal to $\hbar\omega_0$, only the divergent logarithmic terms in the expressions (Al) and (A2) would have to be retained, giving a finite value to γ . But it must be realized that the coefficient d $\ln \delta / d \ln T_c$ in front of the logarithmic term in the expression (A2) is numerically so small that this term makes a non-negligible contribution to D only for values of δ so close to $\hbar \omega_0$ that it would need a precision in the value of the doping ratio x which is experimentally inaccessible. Thus, physically, only the divergent logarithmic term of the numerator of the expression (Al) gives an observable behavior in the variation of γ . Then, using the asymptotic behavior of $K(u_0, u_1)$ given in Appendix A, one gets of formula (15).

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