

## HIGH-TEMPERATURE SUPERCONDUCTIVITY FROM ELECTRON-LATTICE COUPLING

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Within the polaron theory of superconductivity for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  the coupling is considered between the charge carriers and both the stretching mode of the apical oxygens and the sheet-bending mode of the basal-plane oxygens, the latter via the tilting angle of the Cu-oxygen octahedra. The same tilting angle enters the coupling strengths of these lattice vibrations, leading to renormalized frequencies and their oxygen-isotope shifts. It is suggested that the anomalies occurring in the superconducting critical temperature, its isotope shift and the isotope shifts of the lattice frequencies around  $x = 0.12$  are related to the orthorhombic-tetragonal transition of these compounds.

### 1. Introduction. Outline of the Model

The polaron theory of superconductivity<sup>1–5</sup> for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  is used in the present paper to study a certain type of coupling between the charge carriers and the oxygen lattice vibrations, which involves the tilting angle  $\sqrt{\lambda}$  of the Cu-oxygen octahedra. It has been shown<sup>6–8</sup> that the  $x$ -dependence of the critical temperature and its isotope shift can be obtained from such a theory. Recently, certain anomalies have been reported<sup>9,10</sup> in the superconducting critical temperature, its oxygen-isotope shift and the oxygen-isotope shifts of both the stretching mode of the apical oxygens ( $\omega \sim 500 \text{ cm}^{-1}$ ) and the sheet-bending mode of the basal-plane oxygens ( $\omega^* \sim 240 \text{ cm}^{-1}$ ) of these compounds around  $x = 0.12$ . Specifically, on passing through  $x = 0.12$  the critical temperature  $T_c$  exhibits a small indent for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (or, even, vanishes for  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$ ), its isotope shift  $\xi$  displays a sharp decrease of about 0.6, while the isotope shifts  $\eta$  and  $\eta^*$  of the two lattice frequencies mentioned above have a sudden increase toward 1/2. Corroborated with the anomalous behaviour of other physical quantities this piece of information led to the suggestion of a new, distinct, low-temperature tetragonal phase of these compounds.<sup>11,12</sup>

The softening of the tilting mode at low temperatures<sup>13,14</sup> produces in the orthorhombic phase of these compounds a staggered tilting of the Cu-oxygen

octahedra of angle  $\sqrt{\lambda}$ . According to experimental and theoretical evidence<sup>9,15</sup> we assume two types of local vibrations of the Cu-oxygen octahedra as being relevant for the present mechanism of superconductivity. The first is the stretching mode of the apical oxygens. It is described by a set of harmonic oscillators labelled by the Cu-site  $i$ , with relative coordinate  $Q_i$ , (bare) frequency  $\omega_0$  and (relative) mass  $M/2$ ,  $M$  being the oxygen mass. The second type of vibrations correspond to the sheet-bending of the basal-plane oxygens, being described by a set of harmonic oscillators labelled by the sites  $\alpha$  of the basal-plane oxygens, with coordinate  $q_\alpha$ , (bare) frequency  $\omega_0^*$  and mass  $M$ . These types of vibrations bring the main contributions to the transverse-optical phonons of frequencies  $\omega \sim 500 \text{ cm}^{-1}$  and  $\omega^* \sim 240 \text{ cm}^{-1}$ , respectively.<sup>9,10</sup> Due to their axial symmetry these vibrations are expected to couple, most significantly, to the charge carriers in the Cu-oxygen sheets. At the same time, owing to the same axial symmetry, these normal modes of the Cu-oxygen octahedra couple to the Cu-cations in the tilted octahedra by their parallel-to-plane components  $\sqrt{\lambda} Q_i$  and  $\sqrt{\lambda} q_\alpha$ , respectively, where  $\sqrt{\lambda}$  is the tilting angle. This type of coupling may also arise for any other normal modes described by a set of harmonical oscillators labelled by  $i$ , with elastic force constant  $K_0$ , coordinate  $\bar{Q}_i$  and mass  $m$ , where  $\mu = \sqrt{M/m}$  is assumed to be much lesser than unity. Therefore, we assume that the model Hamiltonian of the lattice reads

$$H_{\text{latt}} = \sum_i \left( \frac{1}{M} P_i^2 + \frac{1}{4} M \omega_0^2 Q_i^2 \right) + \sum_\alpha \left( \frac{1}{2M} p_\alpha^2 + \frac{1}{2} M \omega_0^{*2} q_\alpha^2 \right) + \sum_i \left( \frac{1}{2M} \bar{P}_i^2 + \frac{1}{2} K_0 \bar{Q}_i^2 \right) + C \sqrt{\lambda} \sum_i Q_i \bar{Q}_i + C^* \sqrt{\lambda} \sum_{(i\alpha)} q_\alpha \bar{Q}_i, \quad (1)$$

where  $C$  and  $C^*$  are coupling constants and  $(i\alpha)$  denotes the nearest-neighbour Cu-oxygens in the basal plane.

Some uncertainties still persist as regards the electronic structure of the high-temperature superconductors.<sup>16</sup> It seems that a gradual charge transfer would take place in these compounds from the Cu-sites toward the basal-plane oxygen orbitals on increasing  $x$ ,<sup>17-19</sup> the electron spectroscopy experiments<sup>20</sup> indicating that the charge carriers are holes in the oxygen- $p$  orbitals. It is assumed in the present model that the (bare) one-particle states of the charge carriers (holes) are placed on the Cu-sites,<sup>21</sup> their energy band being given by the tight-binding approximation, not for very low values of  $x$ , where the magnetic correlations may be neglected. After including the interaction one arrives at the conclusion that a gradual delocalization toward basal-plane oxygens there occurs with increasing  $x$ , suggesting a strong hybridization between the in-plane Cu- $d$  and oxygen- $p$  orbitals.<sup>16-22</sup> The electronic Hamiltonian is therefore given by<sup>1-5</sup>

$$\mathcal{H}_{\text{el}} = -t_0 \sum_{(ij)\sigma} c_{i\sigma}^\dagger c_{j\sigma} + U_0 \sum_{i\sigma} n_{i\sigma} n_{i-\sigma} + V \sum_{(ij)\sigma\sigma'} n_{i\sigma} n_{j\sigma'}, \quad (2)$$

where  $t_0$  is the bare bandwidth parameter,  $U_0$  and  $V$  are the bare on-site and inter-site Coulomb repulsions, respectively, and  $n_{i\sigma}$  is the site occupancy of the one-electron state with spin  $\sigma$ . As usual in the polaron theory the interaction of the charge carriers with the local vibrations mentioned above

$$\mathcal{H}_{\text{int}} = g \sum_{i\sigma} Q_i n_{i\sigma} + G\sqrt{\lambda} \sum_{(i\alpha)\sigma} q_\alpha n_{i\sigma} , \tag{3}$$

includes, besides the contribution of the apical oxygens (coupling constant  $g$ ), the contribution brought by the basal-plane oxygens (coupling constant  $G$ ); the latter involves the parallel-to-plane component  $q_\alpha\sqrt{\lambda}$  of the vibration coordinate, thus explaining the presence of the tilting angle  $\sqrt{\lambda}$  in the second term of (3). We mention that the softening of the tilting mode at sufficiently low temperatures makes the tilting angle  $\sqrt{\lambda}$  a time-independent (non-fluctuating) parameter, i.e., a non-dynamical variable. In this respect  $\sqrt{\lambda}$  is regarded as the order parameter of the orthorhombic phase.<sup>23,24</sup>

The  $g$ - and  $G$ -couplings can easily be eliminated by a standard canonical transformation generated by

$$S = -\frac{2g}{M\omega_0^2} \sum_{i\sigma} P_i n_{i\sigma} - \frac{G\sqrt{\lambda}}{M\omega_0^{*2}} \sum_{(i\alpha)\sigma} p_\alpha n_{i\sigma} + \frac{2gC\sqrt{\lambda}}{K_0 M\omega_0^2} \sum_{i\sigma} \bar{P}_i n_{i\sigma} , \tag{4}$$

which involves the following displacements of the equilibrium positions of oxygen vibrations:

$$\begin{aligned} Q_i &\rightarrow Q_i - \frac{2g}{M\omega_0^2} \sum_{\sigma} n_{i\sigma} , \\ q_\alpha &\rightarrow q_\alpha - \frac{G\sqrt{\lambda}}{M\omega_0^{*2}} \sum_{(i\alpha)\sigma} n_{i\sigma} , \end{aligned} \tag{5}$$

the last summation being extended over all the Cu-sites which are the nearest-neighbours of the oxygen-site  $\alpha$ . For electronic scale energy much lower than the lattice frequencies the effect of the canonical transformation (4) on the transfer term in the electronic Hamiltonian amounts to dressing up the bandwidth parameter

$$\begin{aligned} t_0 \rightarrow t = t_0 \exp &\left[ -2 \left( \frac{g}{M\omega_0^2} \right)^2 \langle (P_i - P_j)^2 \rangle - \frac{\lambda}{2} \left( \frac{G}{M\omega_0^{*2}} \right)^2 \right. \\ &\times \left. \left\langle \left( \sum_{(i\alpha)} p_\alpha - \sum_{(j\alpha)} p_\alpha \right)^2 \right\rangle - 2\lambda \left( \frac{gC}{K_0 M\omega_0^2} \right)^2 \langle (\bar{P}_i - \bar{P}_j)^2 \rangle \right] , \end{aligned} \tag{6}$$

where the averaging is carried out over the lattice ground-state and does not depend on the nearest-neighbouring sites  $i$  and  $j$ .

One may conclude this section by saying that the lattice Hamiltonian (1) remains unchanged under the canonical transformation (4), while the electronic Hamiltonian (2), (3) is transformed into an effective one,

$$\mathcal{H}_{el} = -t \sum_{(ij)\sigma} c_{i\sigma}^{\dagger} c_{j\sigma} + U \sum_{i\sigma} n_{i\sigma} n_{i-\sigma} - \left( \frac{\lambda G}{2M\omega_0^2} - V \right) \sum_{(ij)\sigma\sigma'} n_{i\sigma} n_{j\sigma'}, \quad (7)$$

where  $t$  is given by (6) and

$$U = U_0 - \frac{g^2}{M\omega_0^2} (1 + 2\lambda C^2/K_0 M\omega_0^2) - \frac{2\lambda G^2}{M\omega_0^{*2}} \quad (8)$$

to the lowest order in  $\lambda$ .

## 2. The Lattice Frequencies and their Isotope Shifts

The  $C$ -coupling in the lattice Hamiltonian (1) is removed by the canonical transformation

$$\begin{cases} \tilde{Q}_i &= Q_i \cos \theta - (\sqrt{2}/\mu) \bar{Q}_i \sin \theta \\ \tilde{P}_i &= P_i \cos \theta - (\mu/\sqrt{2}) \bar{P}_i \sin \theta, \\ \tilde{\bar{Q}}_i &= \bar{Q}_i \cos \theta + (\mu/\sqrt{2}) Q_i \sin \theta \\ \tilde{\bar{P}}_i &= \bar{P}_i \cos \theta + (\sqrt{2}/\mu) P_i \sin \theta, \end{cases} \quad (9)$$

where  $\tan 2\theta = 2\sqrt{2}\lambda\mu C/M\omega_0^2$ , to the lowest order in  $\mu = \sqrt{M/m}$ . The dressed frequency of the apical oxygen vibrations is then given by

$$\omega^2 = \omega_0^2 [1 + 2\lambda(\mu C/M\omega_0^2)^2] \quad (10)$$

and the elastic force constant  $K_0$  becomes

$$\tilde{K}_0 = K_0 - 2\lambda C^2/M\omega_0^2 + 2\lambda K_0(\mu C/M\omega_0^2)^2. \quad (11)$$

One can see from (10) that the  $\sqrt{\lambda}$ -coupling produces the dressing of the isotope shift of the lattice frequency

$$\eta = -\frac{\partial \ln \omega}{\partial \ln M} = \frac{1}{2} - 2\lambda(\mu C/M\omega_0^2)^2 \quad (12)$$

which goes to 1/2 when  $\lambda \rightarrow 0$ .

The  $C^*$ -coupling in the lattice Hamiltonian (1) is eliminated, to the lowest order in  $\lambda$ , by the canonical transformation

$$\begin{cases} \tilde{q}_\alpha &= (1 - \lambda\varphi^2/4)q_\alpha + (\varphi\sqrt{\lambda}/2\mu) \sum_{(i\alpha)} \bar{Q}_i \\ \tilde{p}_\alpha &= (1 - \lambda\varphi^2/4)p_\alpha + (\mu\varphi\sqrt{\lambda}/2) \sum_{(i\alpha)} \bar{P}_i, \\ \tilde{\bar{Q}}_i &= (1 - \lambda\varphi^2/2)\bar{Q}_i - (\mu\varphi\sqrt{\lambda}/2) \sum_{(i\alpha)} q_\alpha \\ \tilde{\bar{P}}_i &= (1 - \lambda\varphi^2/2)\bar{P}_i - (\varphi\sqrt{\lambda}/2\mu) \sum_{(i\alpha)} p_\alpha, \end{cases} \quad (13)$$

where  $\varphi = -2\mu(C^*/M\omega_0^{*2})$  (to the lowest order in  $\mu$ ) and a number of four oxygens for every Cu-site and two coppers for each oxygen-site have been considered, as nearest-neighbours. The lattice Hamiltonian becomes

$$\mathcal{H}_{\text{latt}} = \mathcal{H}_{1\text{latt}} + \mathcal{H}_{2\text{latt}} + \mathcal{H}_{3\text{latt}}, \tag{14}$$

$$\mathcal{H}_{1\text{latt}} = \sum_i \left( \frac{1}{M} P_i^2 + \frac{1}{4} M \omega^2 Q_i^2 \right), \tag{15a}$$

$$\begin{aligned} \mathcal{H}_{2\text{latt}} = & \sum_{\alpha} \frac{1}{2M} P_{\alpha}^2 + \frac{\lambda}{2M} (\mu C^*/M\omega_0^{*2})^2 \left( \sum_{(\alpha\beta)} + \sum'_{(\alpha\beta)(2)} \right) P_{\alpha} P_{\beta} \\ & + \sum_{\alpha} \frac{1}{2} M \omega_0^{*2} \left[ 1 + 2\lambda (\mu C^*/M\omega_0^{*2})^2 \right] q_{\alpha}^2 \\ & + \lambda (\mu^2 C^{*2}/M\omega_0^{*2}) \left( \sum_{(\alpha\beta)} + \sum'_{(\alpha\beta)(2)} \right) q_{\alpha} q_{\beta}, \end{aligned} \tag{15b}$$

$$\begin{aligned} \mathcal{H}_{3\text{latt}} = & \sum_i \frac{1}{2m} \bar{P}_i^2 + \frac{\lambda}{2m} (\mu C^*/M\omega_0^{*2})^2 \sum_{(ij)} \bar{P}_i \bar{P}_j \\ & + \sum_i \frac{1}{2} K \bar{Q}_i^2 - \lambda (C^{*2}/2M\omega_0^{*2}) \sum_{(ij)} \bar{Q}_i \bar{Q}_j, \end{aligned} \tag{15c}$$

where

$$\begin{aligned} K = & K_0 \left[ 1 - 2\lambda C^2/K_0 M \omega_0^2 + 2\lambda (\mu C/M\omega_0^2)^2 \right. \\ & \left. - \lambda (2\mu C^*/M\omega_0^{*2})^2 - 4\lambda C^{*2}/K_0 M \omega_0^{*2} \right], \end{aligned} \tag{16}$$

and  $\sum'_{(\alpha\beta)(2)}$  stands for the summation over the oxygen next-nearest-neighbours with a Cu-site in-between. In deriving (15b, c) summations of the type

$$\begin{aligned} \sum_{(i\alpha)\sigma} \sum_{(j\alpha)\sigma'} n_{i\sigma} n_{j\sigma'} &= 4 \sum_{i\sigma\sigma'} n_{i\sigma} n_{i\sigma'} + \sum_{(ij)\sigma\sigma'} n_{i\sigma} n_{j\sigma'}, \\ \sum_{(i\alpha)} \sum_{(i\beta)} q_{\alpha} q_{\beta} &= 2 \sum_{\alpha} q_{\alpha}^2 + \left( \sum_{(\alpha\beta)} + \sum'_{(\alpha\beta)(2)} \right) q_{\alpha} q_{\beta}, \end{aligned} \tag{17}$$

have been encountered, which can straightforwardly be performed by direct evaluation.

Using the phonon representation the Hamiltonian (15c) can easily be written as

$$\mathcal{H}_{3\text{latt}} = \sum_k \left[ \frac{1}{2m} \bar{P}_k^{(1)} \bar{P}_{-k}^{(1)} + \frac{1}{2} K(k) \bar{Q}_k^{(1)} \bar{Q}_{-k}^{(1)} \right], \tag{18}$$

where

$$\begin{cases} \overline{Q}_k &= \left[ 1 + \lambda (\mu C^*/M\omega_0^{*2})^2 V_k \right]^{1/2} \overline{Q}_k^{(1)} \\ \overline{P}_k &= \left[ 1 + \lambda (\mu C^*/M\omega_0^{*2})^2 V_k \right]^{-1/2} \overline{P}_k^{(1)} \end{cases}, \quad (19)$$

$V_k = 2(\cos k_x + \cos k_y)$  and

$$\begin{aligned} K(k) &= K_0 \left[ 1 - (2\lambda/K_0)(C^2/M\omega_0^2 + 2C^{*2}/M\omega_0^{*2}) \right. \\ &\quad \left. + 2\lambda(\mu C/M\omega_0^2)^2 - 4\lambda(\mu C^*/M\omega_0^{*2})^2 \right. \\ &\quad \left. - \lambda(C^{*2}/K_0 M\omega_0^{*2})V_k + \lambda(\mu C^*/M\omega_0^{*2})^2 V_k \right], \end{aligned} \quad (20)$$

$k$  being the in-plane wave vector. In the long-wavelength limit this implies an isotope shift

$$\overline{\eta} = -2\lambda \left[ (\mu C/M\omega_0^2)^2 - 2(\mu C^*/M\omega_0^{*2})^2 \right]. \quad (21)$$

By means of a similar technique one obtains from (15b)

$$\mathcal{H}_{2\text{latt}} = \sum_{k\rho} \left[ \frac{1}{2M} p_{\rho k}^{(3)} p_{\rho-k}^{(3)} + \frac{1}{2} M \omega_{\rho}^{*2}(k) q_{\rho k}^{(3)} q_{\rho-k}^{(3)} \right], \quad (22)$$

where

$$\omega^{*2}(k) = \omega_0^{*2} \left\{ 1 + 2\lambda (\mu C^*/M\omega_0^{*2})^2 \left[ 1 + \frac{3}{4} V_k + (-1)^\rho \frac{3}{2} \sqrt{s_k^2 + u_k^2} \operatorname{sgn}(u_k) \right] \right\}, \quad (23)$$

$s_k = 4 \cos(k_x/2) \cos(k_y/2)$ ,  $u_k = \cos k_x - \cos k_y$  and  $\rho = 1$  refers to the oxygens in the corners of the unit cell while  $\rho = 2$  stands for the oxygens in the middle of the unit cell. The vibration coordinates in (22) are obtained by the following series of canonical transformations:

$$\begin{cases} q_{\rho k} &= \left[ 1 + \lambda (\mu C^*/M\omega_0^{*2})^2 s_{\rho k} \right]^{1/2} q_{\rho k}^{(1)} \\ p_{\rho k} &= \left[ 1 + \lambda (\mu C^*/M\omega_0^{*2})^2 s_{\rho k} \right]^{-1/2} p_{\rho k}^{(1)}, \end{cases} \quad (24)$$

$s_{1k} = 2 \cos k_y$ ,  $s_{2k} = 2 \cos k_x$ ;

$$\begin{cases} q_{1k}^{(1)} &= q_{1k}^{(2)} + \frac{\lambda}{2} (\mu C^*/M\omega_0^{*2})^2 s_k q_{2k}^{(2)} \\ p_{1k}^{(1)} &= p_{1k}^{(2)} - \frac{\lambda}{2} (\mu C^*/M\omega_0^{*2})^2 s_k p_{2k}^{(2)} \\ q_{2k}^{(1)} &= q_{2k}^{(2)} + \frac{\lambda}{2} (\mu C^*/M\omega_0^{*2})^2 s_k q_{1k}^{(2)} \\ p_{2k}^{(1)} &= p_{2k}^{(2)} - \frac{\lambda}{2} (\mu C^*/M\omega_0^{*2})^2 s_k p_{1k}^{(2)}, \end{cases} \quad (25)$$

(to the lowest order in  $\lambda$ );

$$\begin{cases} q_{1k}^{(2)} = q_{1k}^{(3)} \cos \psi + q_{2k}^{(3)} \sin \psi \\ q_{2k}^{(2)} = q_{2k}^{(3)} \cos \psi - q_{1k}^{(3)} \sin \psi \end{cases} \quad (26)$$

(and the corresponding relations for the canonical-conjugate momenta) where  $\tan 2\psi = s_k/u_k$ . As one can see from (23) the sheet-bending phonons are weakly dispersive (through  $\lambda$ ) and their frequencies split off at  $k_x = \pm k_y$ . Equation (23) predicts a maximum jump in the long-wavelength frequencies of magnitude  $6\lambda(\mu C^*/M\omega_0^{*2})^2$ . Most likely, the frequency  $\omega^* = 240 \text{ cm}^{-1}$  reported experimentally<sup>9,15</sup> corresponds to the long-wavelength average frequency given by (23)

$$\omega^{*2} = \omega_0^{*2} \left[ 1 + 2\lambda (2\mu C^*/M\omega_0^{*2})^2 \right]. \quad (27)$$

The isotope shift of this mode is given by

$$\eta^* = \frac{1}{2} - 2\lambda(2\mu C^*/M\omega_0^{*2})^2, \quad (28)$$

and one can see that it exhibits the same tendency toward 1/2 for vanishing  $\lambda$ . We remark in addition that, according to (23), the zone-center and zone-edge phonons are correlated by the  $\lambda$ -coupling, as it has been suggested.<sup>9,10</sup>

Having the lattice Hamiltonian diagonalized one can easily calculate the reduction in the bandwidth parameter  $t$  as expressed by (6). For the apical-oxygen contribution one obtains straightforwardly  $\langle P_i^2 \rangle = \langle P_j^2 \rangle = M\omega/4$  and  $\langle P_i P_j \rangle = 0$ , by using (15a). For the remaining contribution we should pass to the phonon representation. To the lowest order in  $\lambda$  one gets, for example,

$$\langle \overline{P}_i^2 \rangle = \sum_k \langle \overline{P}_k^{(1)} \overline{P}_{-k}^{(1)} \rangle = \sqrt{mK_0}/2 \quad (29)$$

and  $\langle \overline{P}_i \overline{P}_j \rangle = 0$ , where use has been made of (18)–(20). The contribution of the basal-plane oxygens can be treated analogously. To the same approximation one obtains, for example,

$$\langle p_{\rho k} p_{\rho' k'} \rangle = (M\omega_0^*/2)\delta_{k,-k'}, \quad (30)$$

so that

$$\left\langle \left( \sum_{(i\alpha)} p_\alpha - \sum_{(j\alpha)} p_\alpha \right)^2 \right\rangle = 3M\omega_0^*, \quad (31)$$

where all the oxygens surrounding the nearest-neighbouring sites  $\langle ij \rangle$  have been taken into account. Thereby, the reduced bandwidth parameter  $t$  given by (6) becomes

$$t = t_0 \exp \left\{ -\frac{g^2}{M\omega_0^3} \left[ 1 + \lambda(\mu C/M\omega_0^2)^2 \right] - \frac{3\lambda G^2}{2M\omega_0^{*3}} - 2\lambda\sqrt{mK_0}(gC/K_0M\omega_0^2)^2 \right\}. \quad (32)$$

To summarize, one may say that a polaronic picture is obtained for the charge carriers, as described by the effective electronic Hamiltonian (7). An intersite attraction proportional to  $\lambda$  is present in (7), which we assume to overcome the intersite Coulomb repulsion  $V$ , as to lead to superconductivity; at the same time the on-site Hubbard repulsion  $U_0$  is assumed to be sensibly reduced by the polaronic binding energy (according to (8)) such as not to destroy the superconductivity; and the effective bandwidth (32) is assumed to be greatly reduced as to allow the attractive interaction (which affects all the available electronic states) to be treated in first approximation without damping and retardation effects. We shall see in the next sections that these assumptions are consistent with the experimental data in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .

### 3. The "Renormalization" of the Theory

The variation of the tilting angle with the filling factor  $x$  is given by (5) as

$$\delta(\sqrt{\lambda}) = \delta\bar{q}_\alpha|a = -\frac{2G\sqrt{\lambda}}{aM\omega_0^{*2}}\delta x, \quad (33)$$

where  $a$  is the distance between the Cu cations and the basal-plane oxygens in the untilted phase, and an average site occupancy  $\bar{n}_{i\sigma} = x/2$  has been assumed. For the present purpose we neglect the oxygen deficiency and the effects of the charge localization by disorder, magnetic correlations, impurities, defects, etc., and assume that the concentration of the charge carriers (holes) is given by the doping level  $x$ , as suggested by the electroneutrality of the chemical formula of the compound.<sup>7</sup> From (33) one obtains

$$\sqrt{\lambda} = \sqrt{\lambda_0} \exp\left(-\frac{2G}{aM\omega_0^{*2}}x\right) \quad (34)$$

and

$$\lambda = \lambda_0 \exp\left(-\frac{4G}{aM\omega_0^{*2}}x\right). \quad (35)$$

The linear approximation to (34) and (35) shows that the tilting angle and its square vanish for  $x_{\text{cr}} = (2G/aM\omega_0^{*2})^{-1}$  and, respectively,  $x_{\text{cr}}/2$ , so that one may write

$$\sqrt{\lambda} = \sqrt{\lambda_0} \exp(-x/x_{\text{cr}}) \quad (36)$$

and

$$\lambda = \lambda_0 \exp(-2x/x_{\text{cr}}). \quad (37)$$

The distance between the Cu cations and the basal-plane oxygens in the tilted phase is therefore given by

$$d_1 = a/\cos\sqrt{\lambda} \cong a(1 - \lambda_0 x/x_{\text{cr}}), \quad (38)$$



which reproduces satisfactorily the experimental  $x$ -dependence  $d_1 = 1.9 - 0.06x$  Å for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ <sup>25</sup> for  $a = 1.9$  Å and  $\lambda_0 = 0.007$  (tilting angle  $\sqrt{\lambda_0} \cong 4.9^\circ$ ) provided  $x_{\text{cr}} = 0.23$  as indicated by the orthorhombic-tetragonal transition.<sup>26-28</sup> Having known  $x_{\text{cr}}$  and  $a$  one can easily obtain  $G/M\omega_0^* = a/2x_{\text{cr}} = 4.13$  Å, whence for  $\omega_0^* = 240$  cm<sup>-1</sup> (30 meV) one gets the polaronic coupling strength  $G = 14.3$  eV/Å corresponding to the basal-plane oxygens. We remark that, according to (7), the superconducting strength  $J = \lambda_0 G^2/2M\omega_0^{*2} = 207$  meV is the product of the very small tilting angle square  $\lambda_0 = 0.007$  by the very large polaronic binding energy  $G^2/2M\omega_0^{*2} = 29.6$  eV. We emphasize that the orthorhombic-tetragonal transition is a continuous transition in this model,<sup>24</sup> described by the exponentially descending  $x$ -dependence of the tilting angle. Therefore, if one assumes, according to the experimental evidence, that the tilting angle practically vanishes for  $x_{\text{cr}} = 0.23$  it follows that its square, which decreases at a twice faster rate, (and the orthorhombicity parameter) vanishes around  $x_{\text{cr}}/2 \cong 0.12$ , i.e., exactly where the anomalies occur in these compounds. In addition, the exponential  $x$ -dependence of the tilting angle shows that the superconductivity might be present, as it is, even in the tetragonal phase, where the experiments report a vanishing tilting angle.

The experimental  $x$ -dependence of the tilting angle  $\sqrt{\lambda}$ , which decreases linearly with increasing  $x$  and vanishes for  $x_{\text{cr}} = 0.23$ , suggests that  $G$  in (34) increases with increasing  $x$  and tends to infinity for  $x$  approaching  $x_{\text{cr}}$  (i.e., for vanishing tilting angle). The same conclusion may also be drawn from the linear  $x$ -dependence observed experimentally for the square  $\lambda$  of the tilting angle (35) (and the distance  $d_1$  given by (38)), which suggests that  $G$  tends to infinity for  $x$  approaching  $x_{\text{cr}}/2 \cong 0.12$ . This is also supported by the presence of the superconductivity for values of  $x$  greater than  $x_{\text{cr}}$ , where the superconductivity strength  $J = \lambda G^2/2M\omega_0^{*2}$  must therefore be non-vanishing. In agreement with this evidence the results of the present model are "renormalized" by letting  $\lambda$  go to zero and  $G$  go to infinity, while preserving  $J$  different from zero and independent of  $x$ . A gradual transfer of the charge carriers might be responsible for this enhancement of  $G$ , as already suggested.<sup>16-20</sup> In addition, this trend of the charge carriers to localize on the oxygen-sites diminishes the coefficient of the  $\lambda G^2$ -term in  $t$  given by (32). Therefore, we also make the substitution

$$t_0 \exp(-3\lambda G^2/2M\omega_0^{*3}) \rightarrow \tilde{t}, \quad (39)$$

and treat the "renormalized" bandwidth parameter  $\tilde{t}$ , as well as the superconducting strength  $J$ , as fitting parameters.

Using  $\lambda$  given by (37) one can fit the isotope shifts (12) and (28) of the phonon frequencies to the experimental values  $\eta = 0.44$  and  $\eta^* = 0.38$ , corresponding to  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  for  $x = 0.15$ .<sup>9,10</sup> For  $\lambda_0 = 0.007$  and  $x_{\text{cr}} = 0.23$  established above, and  $\omega_0 = 500$  cm<sup>-1</sup> (62.5 meV),  $\omega_0^* = 240$  cm<sup>-1</sup> (30 meV) one obtains

$\mu C \cong 60 \text{ eV/\AA}^2$  and  $\mu C^* \cong 9.5 \text{ eV/\AA}^2$ . The curves  $\eta(x)$  and  $\eta^*(x)$  obtained for those parameter values are plotted in Fig. 1 and compared with the experimental data<sup>9,10</sup> included also in Table 1. One can see from Fig. 1 that the increasing of  $\eta$  and  $\eta^*$  toward 1/2 is qualitatively accounted by  $\eta(x)$  and  $\eta^*(x)$  obtained above, except for the jump at  $x = 0.12$ .

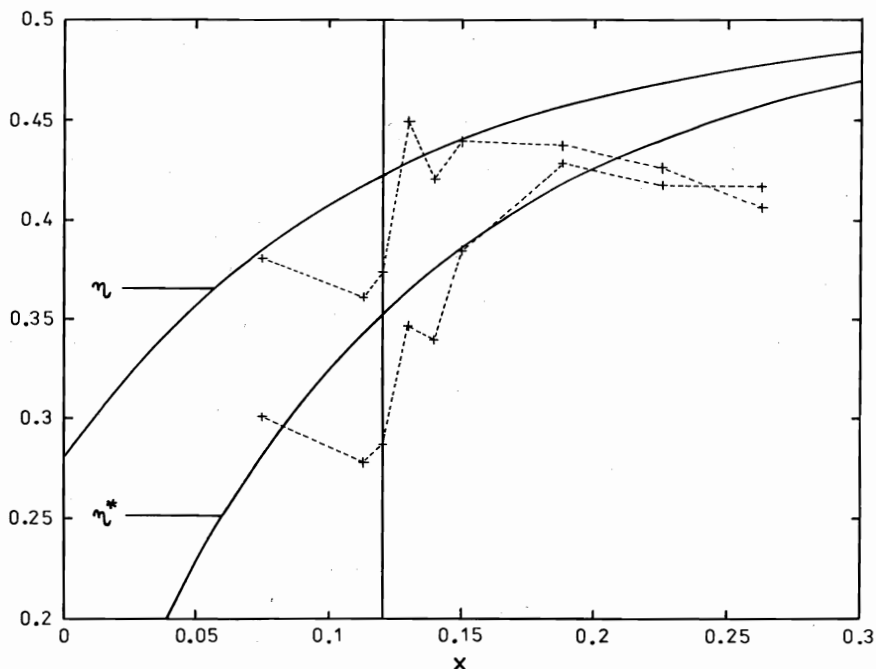


Fig. 1. The isotope shifts  $\eta$  and  $\eta^*$  of the lattice frequencies  $\omega = 500 \text{ cm}^{-1}$  and  $\omega^* = 240 \text{ cm}^{-1}$ , respectively, versus Sr context  $x$  of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ . The solid lines represent the theoretical values (12) and (28) fitted to the experimental data  $\eta = 0.44$  and  $\eta^* = 0.38$  (corresponding to  $x = 0.15$ ) for  $\mu C = 60 \text{ eV/\AA}^2$  and  $\mu C^* = 9.5 \text{ eV/\AA}^2$ . The  $x$ -dependence is contained in the tilting angle given by (35). The crosses represent the experimental data taken from Refs. 9 and 10 (M. K. Crawford *et al.*, *Phys. Rev.* **B41**, 282 (1990); *idem*, *Science* **250**, 1390 (1990)) and included in Table 1. The vertical line indicates the jump at  $x = 0.12$ .

One may expect therefore a sudden decrease  $\Delta\lambda$  at  $x = 0.12$ , which would indicate structural changes accompanying the electronic ones; consequently,  $\Delta G$  and  $\Delta J$  variations are also to be expected at  $x = 0.12$ . Indeed, within the linear approximation  $\lambda$  given by (37) becomes

$$\lambda \cong \lambda_0(1 - 2x/x_{\text{cr}}), \quad (40)$$

which shows that  $\lambda$  vanishes for  $x = x_{\text{cr}}/2 \cong 0.12$ , causing thereby the trend toward 1/2 in  $\eta$  and  $\eta^*$ . Therefore, we "renormalize" the lattice part of the theory by letting  $\lambda$  go to zero and replacing the bare phonon frequencies  $\omega_0$  and  $\omega_0^*$  and their

Table 1. The critical temperature, the relative variations of the oxygen mass and of the lattice frequencies  $\omega = 500 \text{ cm}^{-1}$  and  $\omega^* = 240 \text{ cm}^{-1}$ , the isotope shifts  $\eta$  and  $\eta^*$  of these lattice frequencies and the isotope shift  $\xi$  of the critical temperature for various Sr content  $x$  of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ . The data are taken from Refs. 9 and 10 (M. K. Crawford, *et al.*, *Phys. Rev.* **B41**, 282 (1990); *idem*, *Science* **250**, 1390 (1990)). The numbers in parentheses are the theoretical values obtained in the present work.

$x$	0.075	0.113	0.12	0.13	0.14	0.15	0.188	0.225	0.263
$T_c[\text{K}]$	21.2 (28.5)	29.6 (35.3)	25.4 (36.3)	29.8 (37.7)	32.4 (37.7)	37.8	34.4 (35.1)	23 (27.9)	8 (14.5)
$\delta \ln M$	0.113	0.108	0.115	0.111	0.113	0.109	0.105	0.110	0.108
$-\delta \ln \omega$	0.043	0.039	0.043	0.050	0.048	0.048	0.046	0.047	0.044
$-\delta \ln \omega^*$	0.034	0.030	0.033	0.038	0.038	0.042	0.045	0.046	0.045
$\eta$	0.381	0.361	0.374	0.450	0.421	0.440	0.438	0.427	0.407
$\eta^*$	0.301	0.278	0.287	0.347	0.340	0.385	0.429	0.418	0.417
$\xi$	0.40 (0.31)	0.60 (0.46)	0.78 (0.39)	0.57 (0.04)	0.15 (0.15)	0.08	0.06 (0.07)	0.10 ( $\sim 0$ )	0.12 (-0.4)

isotope shifts by the corresponding dressed quantities measured experimentally. In addition, a decrease  $\Delta J$  is assumed for the superconducting strength  $J$  at  $x = 0.12$ , as remarked above. We note that the dressed phonon frequencies given by (10) and (27) are related to their isotope shifts (12) and (28) by

$$\omega^2 = \omega_0^2(3/2 - \eta), \omega^{*2} = \omega_0^{*2}(3/2 - \eta^*), \quad (41)$$

so that the jump in  $\eta$  and  $\eta^*$  on passing through  $x = 0.12$  might be compensated by corresponding variations in the elastic force constants (included in  $\omega_0$  and  $\omega_0^*$ ), such as to explain the absence of any noticeable variation with  $x$  of the phonon frequencies.<sup>9,10</sup> This again supports the conclusion that structural changes develop in these compounds on increasing  $x$ , besides the electronic ones.

According to the present "renormalization" procedure the parameters of the electronic Hamiltonian (10) become

$$\begin{aligned} \lambda \frac{G^2}{2M\omega_0^2} &\rightarrow \frac{\lambda G^2}{2M\omega^{*2}} = J, \\ U &= \tilde{U} - J_0, \\ \tilde{U} &= U_0 - 4J, \\ t &= \tilde{t} \exp(-J_0/\omega), \end{aligned} \quad (42)$$

where  $U$ ,  $t$  and  $\tilde{t}$  are given by (8), (32) and, respectively, (39) and  $J_0 = g^2/M\omega^2$ . The jump in  $J$  around  $x = 0.12$  induces corresponding variations in the "renormalized" Coulomb repulsion  $\tilde{U}$  and polaron bandwidth  $\tilde{t}$  given by

$$\Delta \tilde{U} = -4\Delta J, \quad \Delta \ln \tilde{t} = -\ln(t_0/\tilde{t}) \cdot \Delta \ln J. \quad (43)$$

We note that on replacing the bare phonon frequencies by their "renormalized" values the dependence of the electronic parameters on the oxygen mass  $M$  occurs through the isotope shifts of the phonon frequencies. Indeed, from  $J_0 = g^2/M\omega^2$  one obtains

$$\frac{\partial \ln J_0}{\partial \ln M} = 2\eta - 1 \quad (44)$$

and, similarly, from (39) and (42) one gets

$$\begin{aligned} \frac{\partial J}{\partial \ln M} &= (2\eta^* - 1) \frac{\lambda G^2}{2M\omega^{*2}} = 0, \\ \frac{\partial \ln \tilde{t}}{\partial \ln M} &= -(3\eta^* - 1) \ln(t_0/\tilde{t}) = -\frac{3}{2} \ln(t_0/\tilde{t}), \end{aligned} \quad (45)$$

where it has been taken into account that  $2\eta^* - 1 = \mathcal{O}(\lambda)$  from (28), and the calculations are carried through to the lowest order in  $\lambda$ .

To conclude this section we note that the distance between the Cu cations and the apical oxygens is given from (5) by

$$d_2 = \text{const} - \frac{g}{M\omega^2} x, \quad (46)$$

the factor 2 in (5) being lost since  $Q_i$  is the relative oxygen-oxygen coordinate. The experimental data<sup>28,29</sup> indicate a slope of 0.1 Å for this linear  $x$ -dependence of  $d_2$ , whence one gets  $g = 1.5 \text{ eV}/\text{Å}$  for  $\omega = 500 \text{ cm}^{-1}$  (62.5 meV). Therefore,  $J_0 = g^2/M\omega^2 = 150 \text{ meV}$ , which is consistent with the polaronic binding energy (corresponding to the apical oxygens) suggested by photo-induced infrared spectroscopy ( $2J_0 \sim 0.3\text{--}0.4 \text{ eV}$ ).<sup>30</sup> In addition, it is worth noting that an oxygen-isotope shift is predicted by the present model for the Cu-oxygen distances  $d_1$  given by (38) and  $d_2$  given by (46) as well as for the tilting angle (36), which might be observed experimentally.

#### 4. The Superconductivity

The electronic Hamiltonian (7), whose parameters are given by (42), has been obtained by assuming that the effect of the electron-lattice interaction on the electronic transfer term is described satisfactorily by averaging this interaction over the ground state of the lattice. Consequently, an effective polaronic bandwidth is obtained, which is much lower than the lattice frequencies, and an electron-electron attraction which acts over all the available electronic states. This is consistent with the reduced effectiveness of the screening in high-temperature superconductors. We assume further that the polaron attraction appearing in (7) may lead to a classical BCS-type of superconductivity via Cooper pairs, the only differences being that we have a reversed scale of energies: the electronic bandwidth is much lower than the lattice frequencies. Consequently, the attractive interaction is not restricted to a thin Debye shell around the Fermi surface but, instead, an electronic cut-off of the

order of the Fermi energy should be used for the pairing processes. We neglect the damping and the retardation effects at this level of approximation, which is consistent with the assumption under which the electronic Hamiltonian (7) has been derived. Assuming a much lower electronic bandwidth than the lattice frequencies one has to worry about the bipolaronic instability in this system. However, taking into account corrections to the electronic transfer term due to the deviation of the electron-lattice interaction from its lattice-averaged value, the electronic bandwidth increases and the occurrence of the bipolaronic instability gets more unlikely. We assume, therefore, that the electronic pairs are Cooper pairs and not bipolarons. However, we note that their radius is of the order of the reciprocal of the Fermi wave vector, and, consequently, much lower than the radius of the classical Cooper pairs. Under these circumstances we further assume that, for low values of the filling factor  $x$ , the Fermi sea is isotropic and the energy levels are given by  $E_k = -2t(\cos k_x + \cos k_y) \cong -4t(1 - k^2/4)$  (according to (7)); therefore, the Fermi wave vector is given by  $k_F = (2\pi x)^{1/2}$ .

The pairing potential (for singlet superconductivity) can be derived from (7) as

$$\begin{aligned} V_{kk'} &= 2(J - V)(\cos k_x \cos k'_x + \cos k_y \cos k'_y) - U \\ &\cong 4(J - V)(1 - k^2/4 - k'^2/4) - U \end{aligned} \tag{47}$$

Under the condition discussed above this potential may be replaced by its value averaged over the Fermi sea

$$\overline{V_{kk'}} = 2\pi(J - V)(\beta - x) , \tag{48}$$

where

$$\beta = \frac{2}{\pi} \left[ 1 - \frac{U}{4(J - V)} \right] . \tag{49}$$

The superconducting critical temperature  $T_c = \beta_c^{-1}$  is then given by the classical BCS-equation

$$\frac{\alpha}{\beta - x} = \int_0^{\pi t x \beta_c} dy \frac{\tanh y}{y} , \tag{50}$$

where

$$\alpha = 2t/(J - V) \tag{51}$$

and the electronic cut-off  $tk_F^2$  has been used, as discussed above. We note that the critical temperature given by (50) is not limited by the screening effects, this being the reason (together with relatively large values of the pairing strength, as we shall see below) for high-temperature superconductivity. However, the damping and the retardation effects may play an important role in lowering the superconducting critical temperature. Equation (50) predicts a non-trivial dependence of the critical temperature on the filling factor  $x$ . This comes, on one hand, from using the electronic cut-off for the pairing processes and, on the other, from the weakening

of the pairing potential with increasing  $x$ , as shown by (48). Indeed, for increasing values of the filling factor  $x$  the pairing processes with large-momentum transfer dominate and the pairing potential (47) acquires, in fact, negative values. This salient feature of  $V_{kk'}$  is incorporated in its approximation given by (48).

The isotope shift  $\xi$  of the critical temperature ( $T_c \sim M^{-\xi}$ ) can easily be derived from (50) by making use of the definitions (42), (49) and (51) and by the  $M$ -dependence of these parameters as given by (44) and (45). After straightforward calculations one gets

$$\xi = (1 - 2\eta) \frac{\alpha^2 J_0 \coth(\pi t x \beta_c)}{4\pi t (\beta - x)^2} + \left[ (3\eta - 5/2) \frac{J_0}{\omega} + \frac{3}{2} \ln(t_0/t) \right] \cdot \left[ 1 - \frac{\alpha}{\beta - x} \coth(\pi t x \beta_c) \right]. \quad (52)$$

One can see that the isotope shift  $\xi$  depends in a complicated way on the model parameters, the isotope shifts of the lattice frequencies, the filling factor and the critical temperature. For  $\eta = 1/2$ , i.e., for a non-interacting harmonical-oscillator mode of the lattice ( $\omega \sim M^{-1/2}$ ),  $\xi$  may acquire low values, either positive or negative, due to the differences included in the square brackets in (52). On the contrary, for  $\eta$  departing from  $1/2$  the  $(1-2\eta)$ -term in (52) brings the main contribution to  $\xi$ .

The experimental  $x$ -dependence of the critical temperature  $T_c$  and its isotope shift  $\xi$  for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4^{9,10}$  given in Table 1 is analyzed by using (50) and (52). In order to fix the values of the parameters these formulae are fitting to the experimental values  $T_c = 37.8$  K,  $\xi = 0.08$  and  $\eta = 0.44$  corresponding to  $x = 0.15$ , for which the experimental data are most accurate. We note that the maximum experimental errors are  $\pm 0.02$  for  $x$  and  $\pm 0.03$  for  $\xi$ , especially for those regions where the critical temperature has a steep  $x$ -dependence.<sup>9,10</sup> Using  $J_0 = 150$  meV, as established in the preceding section,  $\omega = 500$  cm<sup>-1</sup> (62.5 meV) and  $t_0 = 0.6$  eV, corresponding to a bare electron bandwidth  $\sim 5$  eV as indicated by the electronic structure calculations for the present band model,<sup>16,22</sup> the experimental data mentioned above are fitted by (50) and (52) for  $t = 2.4$  meV. One can see that the effective polaronic bandwidth parameter  $t$  is much lower than the lattice frequency  $\omega$ , in agreement with the basic assumption of the model. In addition the effective bandwidth parameter corresponding to the polaron effect of the basal-plane oxygens is  $\tilde{t} = 26.5$  meV, as obtained from (42). We emphasize that the values obtained here for both  $t$  and  $\tilde{t}$  are strongly dependent on the model approximation of averaging the electron-lattice interaction over the lattice ground-state in the electron transfer term (electronic Hamiltonian given by (7)). Higher-order corrections to this approximation, including damping and retardation effects as discussed above, would increase sensibly both the  $t$  and  $\tilde{t}$  values. In addition, on increasing temperature the tilting angle square  $\lambda$  diminishes<sup>24</sup> and, therefore, the parameter  $\tilde{t}$  evolves rapidly toward  $t_0$  as shown by (39). Consequently, the bandwidth parameter of the normal state would rather correspond to  $t_0 \exp(-J_0/\omega) \cong 54$  meV, (i.e., a bandwidth of  $\sim 0.4$  eV, according to our band model). This value of the bandwidth

parameter is consistent with  $\sim 36$  meV, which may be derived within the present band model from the density of states  $\sim 30$  state/Ry-cell required for describing the normal-state properties.<sup>31-33</sup>

The critical temperature given by (50) possesses a maximum  $T_c^{\max}$  for an optimum value of the hole concentration  $x_0$ . Taking  $T_c^{\max} = 37.8$  K and  $x_0 = 0.15$ , as suggested by the experimental data,<sup>9,10</sup> and using  $t = 2.4$  meV obtained above one gets  $\alpha = 0.05$  and  $\beta = 0.3$  from (50) and  $J - V = 92$  meV,  $U = 192$  meV from (51) and, respectively, (49). Using  $J = 207$  meV obtained in the previous section one finds  $V \cong 115$  meV and  $U_0 \cong 1.2$  eV, the latter parameter being obtained by using (42). The bare on-site repulsion  $U_0 \cong 1.2$  eV agrees with the Cu-site Hubbard repulsion  $\sim 2$  eV obtained within a delocalized-orbital model for the antiferromagnetic correlations in the high-temperature superconductors.<sup>34</sup> One may say that the polaronic interaction employed here is strongly enough to overcome intersite Coulomb repulsions as large as  $\sim 115$  meV and to depress sensibly on-site Coulomb repulsions as large as  $\sim 1.2$  eV, such as to make possible the superconductivity. However, we should also emphasize that the bare Coulomb repulsions obtained here strongly depend on the band model; for a more realistic band model one may expect even an  $x$ -dependence of these parameters.

Having known the parameter values in (50) ( $\alpha = 0.05$ ,  $\beta = 0.3$ ,  $t = 2.4$  meV) the  $x$ -dependence of the critical temperature  $T_c$  is obtained from this equation and plotted (solid line) in Fig. 2. One can see that the theoretical values compare satisfactorily with the experimental ones<sup>9,10</sup> (crosses in Fig. 2), except for the small indent observed in  $T_c$  around  $x = 0.12$ . These data are also included in Table 1. Similarly, the isotope shift  $\xi$  can be computed from (52) for various values of the hole concentration  $x$  and the isotope shifts of the lattice frequencies (included in Table 1), by employing the previously established parameter values  $\alpha = 0.05$ ,  $\beta = 0.3$ ,  $t = 2.4$  meV,  $J_0 = 150$  meV,  $t_0 = 0.6$  eV and  $\omega = 62.5$  meV. The critical temperature in (52) is that given by (50). In doing so, we incorporate in the  $x = 0.12$  jump of  $\xi$  only that part which comes from the corresponding jump in  $\eta$ ; its value is about 0.24. As in the case of the critical temperature we should add the variation  $\Delta\xi$  arising from the decrease  $\Delta J$  in the superconducting strength, as established in the preceding section.

The  $\Delta J$ -variations in both the critical temperature  $T_c$  and its isotope shift can easily be obtained from (50) and, respectively, (52) by making use of (43). Straight-forward calculations yield

$$\Delta \ln T_c = \left\{ \frac{\alpha^2 J (4/\pi - x)}{2t(\beta - x)^2} \coth(\pi t x \beta_c) + \left[ \frac{J_0}{\omega} - \ln(t_0/t) \right] \cdot \left[ 1 - \frac{\alpha}{\beta - x} \coth(\pi t x \beta_c) \right] \right\} \Delta \ln J \quad (53)$$

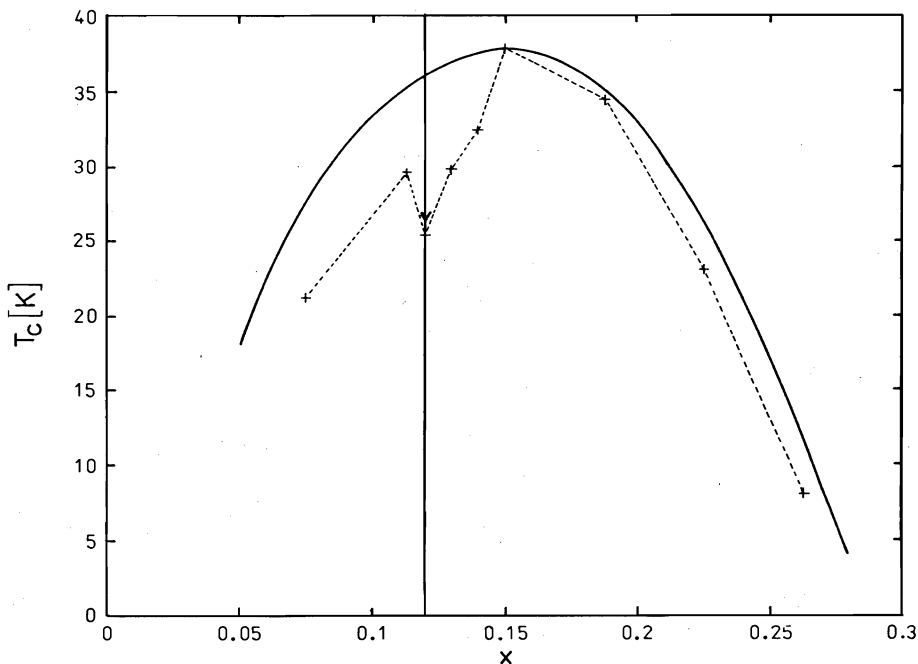


Fig. 2. The critical temperature  $T_c$  vs. Sr content  $x$  of  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  as obtained by fitting (50) to  $T_c^{\text{max}} = 37.8$  K and  $x_0 = 0.15$  for  $t = 2.4$  meV,  $\alpha = 0.05$  and  $\beta = 0.3$ . The crosses indicate the experimental data taken from Refs. 9 and 10 (M. K. Crawford *et al.*, *Phys. Rev.* **B41**, 282 (1990); *idem*, *Science* **250**, 1390 (1990)), included also in Table 1. The arrow on the vertical line at  $x = 0.12$  indicates the decrease in  $T_c$ .

and

$$\begin{aligned} \Delta\xi = & \frac{3}{2} \ln(t_0/t) \left[ 1 + \frac{\alpha}{\beta - x} \frac{\pi t x \beta_c}{\sinh^2(\pi t x \beta_c)} \right] \\ & \cdot \left\{ \Delta \ln T_c + \left[ \ln(t_0/t) - \frac{J_0}{\omega} \right] \Delta \ln J \right\} \\ & + \frac{3}{2} \left[ \ln(t_0/t) - \frac{J_0}{\omega} \right] \cdot \left[ 1 - \frac{\alpha}{\beta - x} \text{contanh}(\pi t x \beta_c) \right] \Delta \ln J, \end{aligned} \quad (54)$$

whence one obtains

$$\Delta \ln T_c \cong 14.5 \Delta \ln J \quad (55)$$

and

$$\Delta\xi \cong 0.38 \Delta \ln T_c, \quad (56)$$

for the parameter values established above and  $x = 0.12$ . The experimental values of the  $T_c$  included in Table 1 for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4^{9,10}$  seem to indicate a decrease  $\Delta \ln T_c \cong 0.3$  around  $x = 0.12$ , which according to (55) and (56), corresponds to



$\Delta \ln J \cong 0.02$ , and implies a decrease  $\Delta \xi \cong 0.11$ . Adding the contribution 0.24 due to the jump in  $\eta$  one gets a total variation in  $\xi$  of about 0.35, which is added to the  $\xi$  values computed from (52) for  $x \leq 0.12$ . The theoretical values of the isotope shift  $\xi$  are plotted as filled circles in Fig. 3 and compared with the experimental values represented by crosses. These data are also included in Table 1. One can see from both Table 1 and Fig. 3 a significant difference between the predicted value of about 0.4 and the experimental value of about 0.8 for the isotope shift  $\xi$  at  $x = 0.12$ . Higher values are obtained from the  $\xi$ -variation around  $x = 0.12$  provided larger bandwidth parameter  $t$  is employed. The tendency of  $\xi$  toward negative values for larger values of  $x$  is also suppressed in this case, in agreement with experimental data. On the other hand, the variation in  $T_c$  around  $x = 0.12$  can be as large as  $\Delta \ln T_c = 1$  (vanishing  $T_c$ ), as in the case of  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4^{10-12}$ ; this implies a total jump in  $\xi$  of about 0.6, which is closer to the experimental value. As it is known, the isotope shift depends strongly on the particular details of the electronic band structure, as it has recently been emphasized,<sup>35</sup> higher values than those predicted by the classical BCS-theory being possible. In general, one should, however, recall that the theoretical values of both  $T_c$  and  $\xi$  obtained here are reliable for  $x$  around 0.15, this being the experimental point which the parameters have been fitted to, as corresponding to the most accurate experimental data. The experimental errors in  $x$  induce a maximum error of about 0.1 in the computed values of  $\xi$ .

## 5. Discussion and Conclusions

The polaron theory of superconductivity<sup>1-5</sup> for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  and  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  has been employed in the present paper in order to study a certain type of coupling between the charge carriers (holes) in the Cu-oxygen sheets of octahedra and the transverse-optical phonons corresponding to the stretching mode of the apical oxygens and the sheet-bending mode of the basal-plane oxygens. This coupling involves the tilting angle  $\sqrt{\lambda}$  of the Cu-oxygen octahedra, and the anomalies reported recently in the  $x$ -dependence of the critical temperature  $T_c$ , its oxygen-isotope shift  $\xi$  and the oxygen-isotope shifts  $\eta$  and  $\eta^*$  of the two lattice modes mentioned above are traced back to the orthorhombic-tetragonal transition of these compounds. Specifically, the increase in the isotope shifts of the phonon frequencies observed around  $x = 0.12$ , as well as the decrease in the critical temperature and its isotope shift at the same  $x$ -value are caused by the vanishing of the tilting angle at  $x_{\text{cr}} = 0.23$  and by the sudden increase in the polaronic strength  $G$  (corresponding to the basal-plane oxygens) associated with this structural transformation. Indeed, the linear approximation to (36) and (37) shows that if  $\sqrt{\lambda}$  vanishes for  $x_{\text{cr}} = 0.23$  then its square  $\lambda$  (which enters both the isotope shifts of the phonon frequencies and the superconducting strength  $J$  given by (42)) vanishes for  $x_{\text{cr}}/2 \cong 0.12$ , i.e., exactly where the anomalies are observed. As superconductivity is present even in the tetragonal phase, the structural transformation implies therefore an increase in the

from (12) as

$$\Delta\lambda = -\frac{1}{2}(M\omega^2/\mu C)^2 \cdot \Delta\eta, \quad (58)$$

whence, having known  $M\omega^2 = 1.5 \text{ eV/\AA}^2$  and  $\mu C \cong 60 \text{ eV/\AA}$  from Sec. 3, and taking  $\Delta\eta \cong -0.07$  from the experimental data in Table 1 one gets  $\Delta\lambda \cong -0.002$ , which means  $\Delta \ln \lambda \cong -0.8$  for  $\lambda = \lambda_0/e$  ( $\lambda_0 = 0.007$ ) at  $x = 0.12$ , according to (37). This  $\Delta\lambda$  variation implies, from (28), a variation  $\Delta\eta^* \cong 0.12$ , which agrees with the experimental data in Table 1. It follows from (57) that an increase of about  $\Delta \ln G \cong 0.39$  occurs in the polaronic coupling strength  $G$  around  $x = 0.12$ . The relatively large, and of opposite signs, variations  $\Delta \ln \lambda$  and  $\Delta \ln G$  reflect the significant changes in both the lattice and the electronic structures of these compounds. The former can be seen, through  $\Delta\lambda$ , in the variation at  $x = 0.12$  of the isotope shifts  $\eta$  and  $\eta^*$ , while both do contribute to the variation of  $\xi$  at  $x = 0.12$  through  $\Delta\eta$  and  $\Delta J$ , as can be seen from (52) and (54). The two large variations of opposite signs  $\Delta \ln \lambda$  and  $\Delta \ln G$  combine to the very small variation  $\Delta \ln J$  for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  thus entailing, according to (53), a small variation  $\Delta \ln T_c$  as compared to  $\Delta\xi$ . Equation (57) shows that the variations around  $x = 0.12$  in  $\eta$ ,  $T_c$  and  $\xi$  are interrelated, via (53), (54) and (58) obtained within the present model. For example, the variation  $\Delta\eta$  (and therefore  $\Delta\lambda$ ) seems to be smaller for  $\text{La}_{2-x}\text{Ba}_x\text{CuO}_4$  as compared with the Sr-doped compound,<sup>10</sup> while the variation  $\Delta T_c$  (and therefore  $\Delta J$ ) is larger, as corresponding to a vanishing critical temperature.<sup>10</sup> Consequently, one may infer from (52) and (54) that  $\Delta\xi$  is smaller in this case, as it seems to be observed experimentally,<sup>10</sup> since  $\Delta J$  brings a smaller contribution to  $\Delta\xi$  as compared to  $\Delta\eta$ . On the other hand, from (57) one may say that  $\Delta G$  is smaller than in the Sr-doped compound, i.e., the changes in the electronic structure are reduced in this case. Not hampered by these changes the lattice structure transformation could be more visible; indeed, a low-temperature tetragonal phase has been reported for the Ba-doped compound,<sup>11,12</sup> not yet found for its Sr-doped counterpart. In this context, the low-temperature tetragonal phase would be marked by non-vanishing tilting angle  $\sqrt{\lambda}$  but a vanishing orthorhombicity parameter (proportional to the square of the tilting angle), which suggests that these structural transformations are continuous.<sup>24</sup>

To conclude, it is worth comparing the values of  $g = 1.5 \text{ eV/\AA}$  and  $G = 14.3 \text{ eV/\AA}$  obtained in Sec. 3 for the electron-lattice coupling constants with the corresponding values of the Coulomb force, as indicated by the interaction Hamiltonian (3). Assuming a charge  $e$  placed at the Cu-site and a charge  $2e$  (corresponding to  $\text{O}^{2-}$ ) placed at the apical oxygen, separated by  $\sim 2.4\text{\AA}$ , one gets  $g \cong 2.5 \text{ eV/\AA}$ ; for the basal-plane oxygens, where the distance is about  $1.9 \text{ \AA}$ , one obtains  $G \cong 8 \text{ eV/\AA}$ . As one can see, these values are of the same order of magnitude as those obtained by the present analysis of the experimental data. The agreement will be more satisfactory if one provides for a static dielectric constant of about 1.7 (which will reduce the  $g$  value from  $2.5 \text{ eV/\AA}$  to  $1.5 \text{ eV/\AA}$ ) and for the in-plane hybridization of the Cu- $d$  and oxygen- $p$  orbitals, which will increase the value of  $G$  to  $14.3 \text{ eV/\AA}$ .

One may conclude therefore that the interaction discussed in the present paper, between the charge carriers and the oxygen-displacive modes of the lattice, operates in these compounds and may lead to high temperature superconductivity.

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