

## Relation between electronic and atomic structure of $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$

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Using a model with strong interatomic correlations previously proposed to explain the electronic structure and pairing in  $\text{CuO}_2$  superconductors, we obtain a staircase behavior of the Fermi level, and a tendency of O(1) ions to order in Cu-O chains. We study the competition between this tendency and long-range Coulomb repulsion, using an Ising model and group-theoretical techniques. Using the cluster variation method, we also study the orthorhombic-tetragonal transition and correlations of the tetragonal phase. All results agree qualitatively with experiments.

### I. INTRODUCTION

One of the more interesting problems raised by the new high- $T_c$  superconductors is the ordering of O atoms in  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  and its relation to superconductivity and other electronic properties. In spite of the considerable research effort, there are important aspects that remain to be clarified. The orthorhombic structures observed for  $x=1$  (OI) (Ref. 1) and  $\frac{1}{2}$ (OII),<sup>2</sup> and the transition to the tetragonal ( $T$ ) phase,<sup>1</sup> have been explained using a model<sup>3</sup> with three arbitrary O-O interactions of the order of  $\sim 0.1$  eV. It is not clear why these interactions should be so small. From Hall measurements<sup>4</sup> the average distance between carriers should be about 7 Å. Thus one expects a large screening length and a nearest-neighbor O-O Coulomb repulsion of the order of the Ry. The facts that it is quite difficult to increase  $x$  from 1 (Ref. 5), that the lattice parameters can be explained in terms of Coulomb energies,<sup>6</sup> and that the only explanation of the observed<sup>7</sup>  $2\sqrt{2} \times 2\sqrt{2}$  phases is given in terms of these energies<sup>8,9</sup> favor also large O-O repulsions.

Hole doping in  $\text{CuO}_2$  planes has been explained, neglecting electronic correlations<sup>10</sup> and assuming that vacancies enter regularly spaced in full Cu-O chains. This assumption contradicts <sup>63</sup>Cu nuclear-quadrupole-resonance (NQR) experiments,<sup>11</sup> which show that for  $x \sim \frac{2}{3}$  the system consists of full and empty chains. Also, there is growing consensus on the fact that correlations are very important in these systems. Varma, Schmitt-Rink, and Abrahams<sup>12</sup> suggested that interatomic correlations are strong. Models based on this idea were used to explain the pairing mechanism<sup>13,14</sup> and electronic structure<sup>15</sup> of  $\text{CuO}_2$  and also  $\text{BiO}_3$  (Refs. 14 and 16) superconductors. In this paper we show that the point of view of strong interatomic interactions provides a consistent physical picture not only of the above-mentioned properties, but also the hole count, ordering of O atoms, and  $O$ - $T$  transition temperature in  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ . We

have performed three different but interdependent calculations: (1) electronic structure of the planes of the O vacancies, using the model of Refs. 12–15, (2) ground-state atomic structure using a model with screened O-O Coulomb interactions,<sup>8,9</sup> modified to take into account the results of (1), and (3)  $O$ - $T$  transition temperature and characterization of the  $T$  phase.

### II. ELECTRONIC STRUCTURE OF THE $\text{CuO}_x$ PLANES

(1) We consider the two-band Hubbard Hamiltonian of Refs. 12–15 for the chains. Disconnected clusters of up to seven atoms were treated exactly and larger systems were solved in perturbations in the hopping as in Ref. 14. Thus  $H = H_0 + H'$ .  $H'$  is given in Ref. 14 and

$$\begin{aligned}
 H_0 = & \sum_{i \in \text{Cu}(1)} (q_i - 1)[\epsilon_d + (q_i - 2)U_d/2] \\
 & + \sum_{j \in \text{O}(1), \text{O}(4)} (q_j + 2)[\epsilon_p^j + (q_j + 1)U_p/2] \\
 & + U_{pd} \sum_{i \langle j \rangle} q_i q_j, \quad (1)
 \end{aligned}$$

where  $q_k$  is the charge operator on atom  $k$ , the first term describes the energy and Coulomb repulsion between holes added on Cu(1) atoms,<sup>1</sup> the second is the analogous term for O(1) and O(4) atoms in the notation of Ref. 1 [here O(1) is extended to denote any O atoms lying between two Cu(1) atoms], and the last describes the Coulomb repulsion between Cu(1) and neighboring O holes. Equation (1) is written in such a way that the parameters of the model are independent of O(1) atomic occupation (assuming that the interactions with neutral O atoms are negligible). This is not possible with the choice of vacuum of Refs. 13–15.  $H_0$  is independent of the symmetry of the relevant orbitals. In the hopping term  $H'$  we assume that these orbitals are those pointing to the nearest neighbors.

We take  $\epsilon_p^j = \epsilon_p$  for O(1) atoms and  $\epsilon_p^j = \epsilon_p + 2U_{pd}$  for

O(4) atoms due to the repulsion between O(4) holes and Cu(2)<sup>2+</sup> ions.<sup>1</sup> According to Eq. (1) for  $x=1$  (or for isolated CuO<sub>2</sub> planes), the difference between the energies necessary to add a hole in an O(1) or in a Cu ion in the reference state Cu<sup>+</sup>O<sub>2</sub><sup>2-</sup> is

$$\Delta = \varepsilon_p - \varepsilon_d + 10U_{pd}. \quad (2)$$

We expect  $\Delta$  and  $U_{pd}$  to be roughly the same as in CuO<sub>2</sub> planes studied before.<sup>12-15,17</sup> This is supported by band structure calculations<sup>18</sup> in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>.

For  $x=0$ , there are no O(1) atoms. The system consists of O(4)<sup>2-</sup>Cu<sup>+</sup>O(4)<sup>2-</sup> isolated triatomic chains (ITC's). When a neutral O(1) is added, it connects two neighboring ITC's forming a seven-atom cluster. If only O(1)-O(1) Coulomb repulsion is considered, for  $x \leq \frac{1}{2}$ , the O(1) atoms arrange themselves without common Cu neighbors.<sup>8,9</sup> We call these structures "(nearly) hexagonal" structures (HS). They differ from the structures composed of full and empty Cu-O(1) "chain" structures (CS) in the concentration of ITC's, seven-atom clusters, and infinite chains. We have compared the electronic structure for both atomic structures, by diagonalizing exactly the finite systems and perturbatively<sup>14</sup> the infinite chains. Results for reasonable values<sup>17</sup> of the parameters are shown in Fig. 1. The gain in energy for CS,  $\Delta E > 0$ , except for very large values of  $\Delta$  or very small values of  $U_{pd}$ .<sup>19</sup> For  $x < \frac{1}{2}$ ,  $\varepsilon_F$  is determined by the energy necessary to add the first hole on an ITC. For  $x > \frac{1}{2}$  there are no ITC's for HS, and all of them have already one hole for CS. The second plateau in  $\varepsilon_F$  is determined by the energy necessary to add the third hole to a seven-atom cluster for HS and the energy necessary to add the second hole on an ITC for CS. For  $\Delta < 3U_{pd} < 2\Delta$ , the first two holes added to a seven-atom cluster (ITC) are mainly of Cu [O(4)] character, while the third is mainly of O(Cu) character.

So far, the  $2x$  holes per unit cell have been added to the

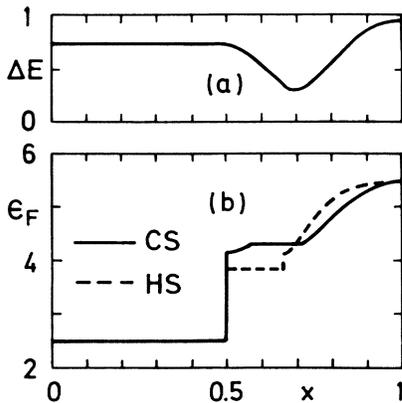


FIG. 1. (a) Difference in electronic energy between "disperse" structures (HS) and "chain" structures (CS) over the difference of second-neighbor O(1) atoms with a Cu in between, and (b) energy necessary to add the next hole, as a function of O content. Parameters are  $U_d, U_p \rightarrow \infty, \Delta/t=3, U_{pd}/t=1.3$ , and  $t'/t=1.28$ , where  $t(t')$  is the Cu-O(1) [Cu-O(4)] hopping. Here terms of order  $tt'$  were neglected in the perturbative calculations for infinite chains (Ref. 19).

"chain" subsystem. According to experiment,<sup>4</sup> with increasing  $x$ , after the discontinuity in  $\varepsilon_F$  at  $x = \frac{1}{2}$ , holes go into the conducting CuO<sub>2</sub> planes until  $n_H \sim 0.24$  holes per unit cell are filled. For CS, from the counting of ITC levels, one realizes that this number should remain approximately constant for  $(n_H + 1)/2 < x < x_c$ , where  $x_c = (2 + n_H)/3 \sim 0.75$ . The agreement with Ref. 4 is good, but that with the corresponding plateau in the superconducting  $T_c$  is even better.<sup>20</sup> For  $x < 0.5$  the system is insulating. For CS the number of Cu<sup>+</sup> varies roughly as  $\sim 1-x$  and the number of O holes as  $\sim x$ , but only holes in CuO<sub>2</sub> planes are conducting, also in agreement with experiment.<sup>21</sup>

### III. GROUND STATE OF THE ATOMIC STRUCTURE

We consider an Ising model for the O(1) sublattice. Any two O ions at a distance  $R_\delta$  repel each other with a screened Coulomb interaction:<sup>8,9</sup>

$$V_\delta = \frac{A}{R_\delta} e^{-R_\delta/\lambda}.$$

However,

$$V_{2Cu} = f \frac{A}{a} e^{-a/\lambda},$$

where  $V_{2Cu}$  is the interaction between second O nearest neighbors with a Cu in between,  $a$  is the lattice parameter (assumed equal to  $b$ ), and  $f \leq 1$  is a reduction factor related to  $\Delta E$  of case (1) by  $\Delta E = (1/f - 1)V_{2Cu}$ . For  $x \geq \frac{1}{2}$ ,  $\Delta E$  varies, and  $\lambda$  and  $A$  decrease with  $x$ , which makes an estimation of the O chemical potential difficult.

We have compared the energies of all structures whose unit cell, or a multiple of it, is a rectangle of largest side  $\leq 3a$  and area  $\leq 8a^2$ . We considered also the unit cells of

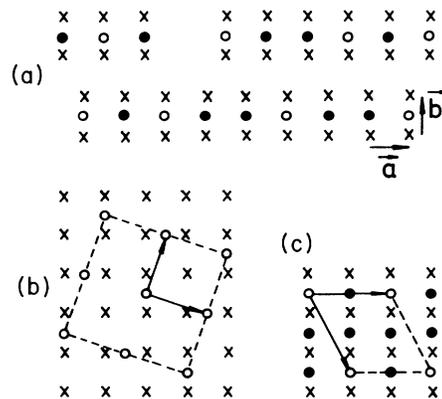


FIG. 2. Different O(1) arrangements. Crosses denote Cu atoms, and for  $x \geq \frac{1}{2}$  ( $x < \frac{1}{2}$ ) the solid (open) circles denote O atoms and the open (solid) circles represent vacancies for  $x=1$ . (a) Unit cells of "chain" structures (CS) for  $x = \frac{1}{2}, \frac{2}{5} (\frac{2}{5}),$  and  $\frac{3}{8} (\frac{3}{8})$ . (b) Structure given by a simple lattice of O atoms for  $x = \frac{3}{4}$ . (c) Structure for  $x = \frac{3}{4}$ , formed by a simple lattice of O vacancies added to the OI structure with  $x=1$ . In (b) and (c), basic vectors (see Table I) and unit cell are shown.

the form  $1 \times n$  with  $n$  up to 8. Based on previous results<sup>8,9</sup> for  $f=1$ , we have also included the simple lattices of O ions [Fig. 2(b)] or vacancies added to the OI,  $x=1$  phase [Fig. 2(c)] in the range  $\frac{1}{8} \leq x \leq \frac{7}{8}$  which were not already considered. All the structures so far observed or proposed are included in this set. We have obtained all structures that are not equivalent by symmetry, using a group-theoretical method developed by one of us,<sup>22</sup> which was modified to take into account the translational symmetry.

In Table I we present results for  $\lambda=a_0$ , where  $a_0=a/\sqrt{2}$ . For  $f=1$  and  $\lambda \sim a_0$ , previous hypothesis<sup>8</sup> and Monte Carlo results<sup>9</sup> are confirmed and only simple lattices (that are in general small deformations of an hexagonal lattice) describe the ground state (GS). For decreasing  $f$ , after a short interval in which other structures appear, the GS for  $x$  near  $\frac{1}{2}$  is dominated by CS and the

TABLE I. Ground-state atomic structures of  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  as a function of  $x$ , for a screening length  $\lambda=a/\sqrt{2}$  and different reduction factors  $f$  of the repulsion between second O nearest neighbors with a Cu ion in between [see Eq. (3)]. For  $f=1$  and  $x \leq \frac{2}{3}$  or  $x=1$ , the O ions are arranged in a simple lattice of the basic vectors given in the second column, with  $\mathbf{c}=(\mathbf{b}+\mathbf{a})/2$ ,  $\mathbf{d}=(\mathbf{b}-\mathbf{a})/2$ . For the other compositions, the structure is formed subtracting a simple lattice of O ions of given basic vectors, from the structure of  $x=1$ . CS ( $1 \times n$ ) denotes ordering in chains with unit cell ( $1 \times n$ ) [see Fig. 2(a)]. PS represents structures formed adding pairs of O atoms with a common nearest-neighbor Cu in between or subtracting them from the  $x=1$  structure. For values of  $x$  not given in the table and  $\frac{1}{8} < x < \frac{7}{8}$ , the system decomposes into the two neighboring structures.

| $f$            | $x$ | 0.7-1         | 0.6                | 0.1                |
|----------------|-----|---------------|--------------------|--------------------|
| $\frac{1}{8}$  |     | 4c, 2c+4d     | idem $f=1$         | idem $f=1$         |
| $\frac{2}{15}$ |     | 4c+d, c+4d    | idem $f=1$         | idem $f=1$         |
| $\frac{1}{7}$  |     | 4c+d, -2c+3d  | idem $f=1$         | idem $f=1$         |
| $\frac{1}{6}$  |     | 2c+3d, -2c+3d | idem $f=1$         | idem $f=1$         |
| $\frac{2}{11}$ |     | 3c+d, -2c+3d  | idem $f=1$         |                    |
| $\frac{1}{5}$  |     | 3c+d, -c+3d   | idem $f=1$         |                    |
| $\frac{2}{9}$  |     | 3c+d, -3d     | idem $f=1$         |                    |
| $\frac{1}{4}$  |     | 3c+d, -2c+2d  | idem $f=1$         | PS                 |
| $\frac{1}{3}$  |     | 2c+2d, c-2d   | idem $f=1$         | CS( $1 \times 3$ ) |
| $\frac{3}{8}$  |     |               |                    | CS( $1 \times 8$ ) |
| $\frac{2}{5}$  |     | 2c+d, c-2d    | idem $f=1$         | CS( $1 \times 5$ ) |
| $\frac{3}{7}$  |     |               |                    | CS( $1 \times 7$ ) |
| $\frac{1}{2}$  |     | 2c, c-2d      | CS( $1 \times 2$ ) | CS( $1 \times 2$ ) |
| $\frac{5}{7}$  |     |               |                    | CS( $1 \times 7$ ) |
| $\frac{3}{5}$  |     |               |                    | CS( $1 \times 5$ ) |
| $\frac{5}{8}$  |     |               |                    | CS( $1 \times 8$ ) |
| $\frac{2}{3}$  |     | c+d, c-2d     | idem $f=1$         | CS( $1 \times 3$ ) |
| $\frac{3}{4}$  |     | 2a, a-2b      | idem $f=1$         | PS                 |
| $\frac{4}{5}$  |     | 2a+b, a-2b    | idem $f=1$         |                    |
| $\frac{5}{6}$  |     | 2a+2b, a-2b   | idem $f=1$         | idem $f=1$         |
| $\frac{7}{8}$  |     | 2a+2b, a-3b   | idem $f=1$         | idem $f=1$         |
| 1              |     | c+d, c-d      | idem $f=1$         | idem $f=1$         |

range of compositions in which this happens grows in general symmetrically around  $\frac{1}{2}$  for decreasing  $f$ . This behavior is observed for all  $\lambda$ . The values of  $f$  for which the transition takes place and the number of different structures increase with  $\lambda$ . For  $\lambda=0.2a_0$ , there are at most nine different GS as  $x$  is varied, and for  $f>0.05$ , there are no CS. For  $\lambda=7a_0$ , CS persist even for  $f=0.8$ . At the transition from CS to other types of ordering, steps in  $\epsilon_F$  and hole occupations are expected according to Sec. II.

For  $x=1$  the GS is the experimentally observed (OI).<sup>1</sup> The same happens for  $x=\frac{1}{2}$  (OII)<sup>2</sup> if  $f$  is sufficiently smaller than 1. For still smaller  $f$ , the obtained GS for  $x=\frac{2}{5}$  and  $\frac{3}{5}$  are consistent with the diffuse streaking seen in Ref. 23 and the same happens for  $x=\frac{2}{3}$ .<sup>24</sup> These structures could only be obtained as metastable with other models.<sup>25,26</sup> The GS for  $x=\frac{1}{8}$  and  $\frac{7}{8}$  and all positive  $f$ , with a unit cell  $2\sqrt{2} \times 4\sqrt{2}$ , is consistent with a unit cell multiple of  $2\sqrt{2} \times 2\sqrt{2}$  required by the experimental observations.<sup>7</sup> The structures proposed in Ref. 7 have a very small energy difference with the GS [ $6.63 \times 10^{-5}$  V for  $\lambda=a_0$ ,  $2.71 \times 10^{-4}$  V for  $\lambda=10a_0$ , where  $V=A/a_0$  is the unscreened O(1) nearest-neighbor interaction]. Thus it is likely that this less-ordered structures are favored except at very low temperatures.

In summary, for  $\lambda \gtrsim 0.4a_0$  and  $f$  small but positive, all observed diffraction patterns<sup>1,2,7,23,24</sup> are explained in terms of the ground state for the corresponding composition ( $x=0, \frac{1}{8}, \frac{1}{2}, \frac{3}{5}, \frac{2}{3}, \frac{7}{8}, 1$ ).

#### IV. THERMODYNAMICS

It remains to explain why the  $T$ -O transition temperature  $T_c$  is much lower than  $V$  (which one expects to be of the order of several eV). We performed a cluster-variation-method (CVM) calculation comparing the free energy of the OI, OII, and  $T$  phases, as in Ref. 3. However, we took our basic figure as a nine-point square cluster since we find that to describe the energy of the important structures for  $\lambda \sim a_0$ , interactions up to at least five nearest neighbors (NN) should be included. Since we are interested in temperatures much lower than the first NN repulsion, we eliminated the configurations with first NN O atoms. Therefore, it was more convenient for us to use the natural iteration method<sup>27</sup> which works with configurational probabilities. However, we solved self-consistently the equations for the constraints establishing a hierarchy of Lagrange multipliers. The method converged for practically all temperatures.

In Fig. 3 a typical evolution of site occupancies is shown.  $T_c$  decreases by a factor of the order of 2 if  $f$  is increased to 0.4.  $T_c$  also decreases strongly with decreasing  $\lambda$ . The low  $T_c$  is due to the fact that the  $T$  phase gains considerable energy avoiding first NN O(1), while the entropy for  $x \sim \frac{1}{2}$  is lowered by only  $\sim 30\%$ .

In Fig. 4 we show the following correlation functions of the  $T$  phase (at temperatures much higher than all interactions except the first NN ones):

$$c_i = \langle (n_j - n_0)(n_{j+\delta_i} - n_0) \rangle / n_0^2. \quad (4)$$

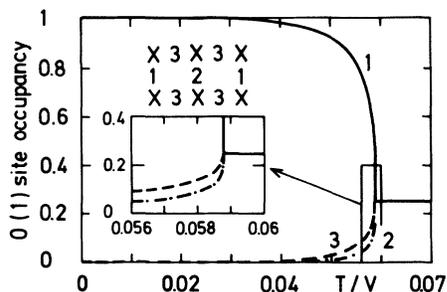


FIG. 3. Site occupancies of the inequivalent sites of the OII phase (Fig. 1, top left) as a function of temperature for constant  $x = \frac{1}{2}$ ,  $\lambda = a_0 = a/\sqrt{2}$ , and  $f = 0$ .  $V = A/a_0$ .

Here  $n_j$  is the occupation number of any site  $j$ ,  $n_{j+\delta_i}$  is that of one of its  $i$ th NN, and  $n_0 = \langle n_j \rangle = x/2$ .  $c_1 = -1$ . The short-range correlations resemble those of the OI phase for all compositions. This agrees with the results of perturbed-angular-correlation spectroscopy.<sup>28</sup> The fact that the  $T$  phase should avoid first NN O atoms was pointed out some time ago by several authors.<sup>5,6</sup>

In summary, covalency of Cu—O bonds and strong in-

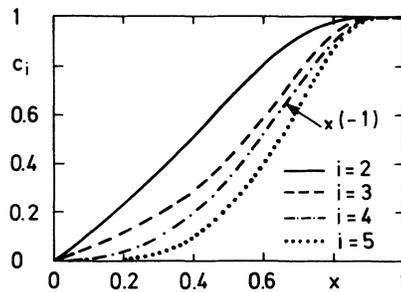


FIG. 4. Correlation functions between  $i$ th neighbors [see Eq. (4)] of a disordered phase without nearest-neighbor O(1) pairs.

teratomic correlations with an intermediate degree of screening can explain the main features of the electronic and atomic structure, thermodynamics and excitonic pairing mechanism<sup>13,14</sup> of  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ .

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