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Electronic-structure calculation of the hole-carrier-density distribution in $Bi_2Sr_2CuO_6$, $Bi_2Sr_2CaCu_2O_8$, and $Bi_2Sr_2Ca_2Cu_3O_{10}$ superconductors

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Results of electronic structure calculations for the hole density in the two-dimensional CuO_2 planes are presented in this paper for the three bismuth cuprate superconductors $Bi_2Sr_2CuO_6$, $Bi_2Sr_2CaCu_2O_8$, and $Bi_2Sr_2Ca_2Cu_3O_{10}$. The hole density in these compounds arises due to electron transfer from the CuO_2 planes to the Bi-O layers. Our calculations show that $Bi_2Sr_2CuO_6$ is so highly overdoped that it is really a metal and at the limit of superconductivity. Also $Bi_2Sr_2CaCu_2O_8$ has a hole density which is higher than is required for a maximum value of T_c. On the other hand, the hole density in the three-layer compound $Bi_2Sr_2Ca_2Cu_3O_{10}$ is nearly optimum, which indicates that apart from the number of layers, an optimum value of the hole density is important for maximizing T_c. The hole densities in the two types of CuO_2 layers in this compound are nearly equal although the central layer has a slightly lower value. As the number of CuO_2 planes increases in this family of superconductors the total amount of hole carrier density per chemical formula increases due to an enhanced electron transfer to the Bi-O layers. However, this increase is small so that the hole density per CuO_2 drops rapidly as the number of CuO_2 planes increases.

I. INTRODUCTION

The bismuth cuprate superconductors are considered to be interesting materials for technological applications. In this family essentially three classes of superconductors have been discovered to date and the superconducting transition temperature T_c depends strongly upon the number of two-dimensional CuO_2 planes per chemical formula.[1-3] For the single CuO_2 layer compound $Bi_2Sr_2CuO_6$ (Bi-2:2:0:1) $T_c < 10$ K but increases to $T_c \sim 80$ K for the two layer compound $Bi_2Sr_2CaCu_2O_8$ (Bi-2:2:1:2), and the highest value $T_c \sim 110$ K is obtained for the three layer compound $Bi_2Sr_2Ca_2Cu_3O_{10}$ (Bi-2:2:2:3) in this family of superconductors. Except for the number of CuO_2 planes, which varies, structurally all these superconductors[4-6] are quite similar (see Fig. 1). In $Bi_2Sr_2CuO_6$ a CuO_2 plane is sandwiched in between two SrO planes and double BiO layers (Bi_2O_2 layers). A Cu atom in this compound is thus octahedrally coordinated by the oxygen atoms. In $Bi_2Sr_2CaCu_2O_8$ there is an extra $CaCuO_2$ layer inserted in between the SrO layers. Structurally this results in two CuO_2 planes separated by a Ca plane in which there are no oxygen atoms. Each Cu atom in this compound is thus pyramidally coordinated by the oxygen atoms. In the third compound with the highest T_c there is an additional $CaCuO_2$ layer, and thus there are three CuO_2 planes sandwiched in between the SrO layers, each of which is separated from the other CuO_2 plane by a Ca plane having no oxygen atoms in it. As shown in Fig. 1, in contrast to the first two compounds where all Cu atoms have similar environments, in the third compound there are quite clearly two types of Cu atoms. The Cu atoms in the central layer have a square planar coordination by the oxygen atoms, while those in the layers above and below are pyramidally coordinated.

It is now well recognized that the superconductivity in the cuprates arises from the presence of the two-dimensional CuO_2 planes. In their ground state they are antiferromagnetic insulators, but on doping by holes or electrons they become metallic and superconducting for a certain range of dopings. Indeed, a direct correlation has been established[7] between the value of T_c and the hole density for the hole doped superconductors, and it has been observed that the T_c varies approximately in an inverted parabolic manner as a function of the hole density in the CuO_2 planes. In the La_2CuO_4 type of compounds, this hole doping is achieved by a direct chemical substitution of, for example, Sr at the La site. In the $YBa_2Cu_3O_{6+\delta}$ family of superconductors, this is done, on the other hand, by varying the stoichiometry in oxygen, and these are thus considered to be self-doped superconductors. For $\delta > 0.4$, ordered CuO chains are formed in this compound. The electrons are transferred from the CuO_2 planes to the CuO chains, which thus act as electron reservoirs for the creation of the hole density in the CuO_2 planes.

In an alternate picture, the occurrence of superconductivity in the cuprates has been associated with a valence of Cu in the CuO_2 planes of greater than +2, since in their antiferromagnetic insulating ground states Cu resides in these compounds with the electronic configuration Cu^{2+}. In this picture, one determines the valence of Cu simply by assuming the nominal ionic

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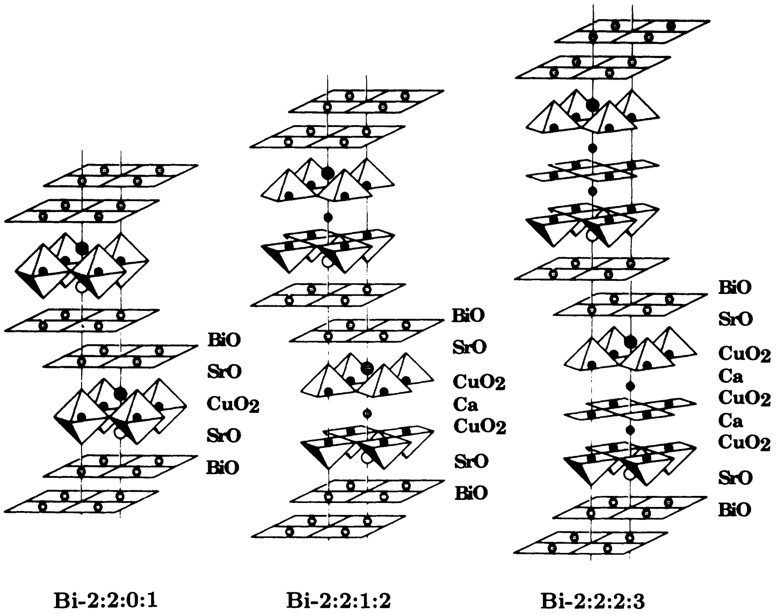

FIG. 1. Crystal structures of $Bi_2Sr_2CuO_6$, $Bi_2Sr_2CaCu_2O_8$, and $Bi_2Sr_2Ca_2Cu_3O_{10}$.

valence of all elements forming the compound. In the bismuth-based cuprate superconductors, assuming Bi, Sr, Ca, and O to be in their ionic states Bi^{3+}, Sr^{2+}, Ca^{2+}, and O^{2-} respectively, one finds Cu to be in its ionic state Cu^{2+}. The bismuth cuprates thus should have been insulating in this ionic picture. Detailed electronic structure calculations[8-11] have, however, shown that the BiO bands cross the Fermi level, E_F, in these compounds, and are therefore coupled to the CuO_2 planes. The Bi_2O_2 layers thus act as electron reservoirs for electron transfer from the CuO_2 planes, and provide the necessary doping.

The anomalously low value of T_c observed for the single layer superconductor Bi-2:2:0:1 could either be due to the fact that it is unreasonably underdoped or highly overdoped. In fact, this compound is quite close to a normal metal and right at the limit of superconductivity. This suggests that it is highly overdoped, and experimental data[12-16] support this idea, since the T_c of this compound can be raised to ~36 K by a partial substitution of La at the Sr site, which will lower the hole density.[16] This seems to be true[17] also for the two-layer compound Bi-2:2:1:2 but to a lower extent, since the T_c in this compound can be raised to ~90 K from the original value of ~80 K by a partial substitution of Y at the Ca site.[18]

The three-layer compound Bi-2:2:2:3 is the most interesting, since it has the highest value of T_c but is also more complicated, since it has two different types of CuO_2 planes in which the hole densities may not be the same. The hole densities in this compound have been estimated by Di Stasio, Müller, and Pietronero[19] in an electrostatic model assuming that in each CuO_2 plane the hole density was uniformly distributed. These authors found that the holes were primarily localized in the two planes with pyramidal coordination of Cu by oxygens, and that the central plane with the square planar coordination contained only ~10 percent of the total hole density. Haines and Tallon[20] improved this calculation by assuming point charges at the Cu and O sites in the CuO_2 planes, and obtained nearly the same hole densities in the two types of CuO_2 planes. Considering this discrepancy in the two calculations, a reliable estimate of the hole density in this compound seems essential, which could lead to an understanding of the role of the interlayer coupling between the CuO_2 planes in raising the value of T_c.

In this paper, the results of our electronic-structure calculations are presented for the three bismuth cuprate superconductors Bi-2:2:0:1, Bi-2:2:1:2, and Bi-2:2:2:3, which allow an estimate of the hole densities to be made in these compounds. Details of the calculations are presented in Sec. II, and Sec. III is devoted to a discussion of the results obtained. Concluding remarks are given in Sec. IV.

II. DETAILS OF CALCULATION

A schematic representation of the crystal structures of the three compounds is shown in Fig. 1. For $Bi_2Sr_2CuO_6$ the crystal structure data was taken from the work of Torardi *et al.*[4] on the stoichiometric compound (orthorhombic space group *Pnan*, $Z=4$) obtained by the substitution of ~10% Pb at the Bi site; this substitution hinders the insertion of additional oxygen atoms into the double Bi-O layers so that this crystal structure does not exhibit the modulations and superstructures seen in the lead free samples. The crystal structure data for $Bi_2Sr_2CaCu_2O_8$ was taken from the work of Calestini *et al.*[5] also with orthorhombic space group *Pnan* ($Z=4$), and again on lead-doped samples, while for $Bi_2Sr_2Ca_2Cu_3O_{10}$ the structural data (orthorhombic space group *A*2*aa*) of Miehe *et al.*[6] was used. The electronic-structure calculations were performed using the tight-binding recursion method with clusters of approximately 12 000 atoms in each compound. The recursion method is inherently advantageous for these compounds, since it is a real-space technique and requires no periodicity, in contrast to the usual *ab initio* reciprocal space methods, which are based on Bloch's theorem and thus require periodicity in the structure. The atomic displacements from the ideal positions in the lattice, which is usual in this class of superconductors, can thus be fully taken into consideration in this method. Further, these compounds have large unit cells, which can be troublesome in achieving the convergence in the calculated energy eigenvalues by the *ab initio* methods. In the recursion method the densities of states are calculated directly at the individual atomic sites through the recursion technique which amounts essentially to an outward integration. A given atom interacts directly with its neighbors, which in turn interact with theirs, leading to an indirect interaction of the central atom with distant neighbors. This technique has been described in detail elsewhere,[21] where a complete mathematical formulation has been given. In our calculations p wave functions at oxygen and bismuth sites and d wave functions at other sites were included. The $2s$ orbitals of oxygens and $6s$ of bismuth lie at much lower energies and thus play no essential role in bonding. The transfer integrals and on-site energies were taken from the work of Hybertsen and Mattheiss,[8] and a $1/d^2$ distance dependence, where d is the interatomic separation, is used to take into account the distance dependence of the hopping integrals. The densities of states obtained with this method[11] are in good agreement with more elaborate *ab initio* calculations[8–10] and demonstrate the reliability of this method and the transferability of the parameters in the study of these compounds.

III. RESULTS AND DISCUSSION

An important crystallographic property of the cuprate superconductors is that the planar copper-apical oxygen bond length in these compounds is ~25% longer than the other Cu—O bond lengths in the crystal. This renders the interaction of the planar Cu atom with the apical oxygen atom much weaker relative to the interaction with the oxygen atoms in its own plane. Thus despite an octahedral or square pyramidal coordination the Cu atom in reality retains a square planar coordination. In fact, the Bi atom forms a natural partner of the apical oxygen atom, since the Bi-apical oxygen atom bond is much shorter. This leads to the formation of two separate sets of band complexes in the vicinity of the Fermi level: (a) planar CuO_2 bands formed between the planar Cu $d_{x^2-y^2}$ orbitals and the planar p_x and p_y oxygen orbitals and (b) BiO_2 bands formed between Bi and the oxygens in its own plane and the apical oxygen. The charge transfer from the CuO_2 planes depends upon the relative positions of the planar CuO_2 bands with respect to the BiO_2 bands.

In Table I we have given the effective charges at different atomic sites in the three compounds calculated by integrating the densities of states at these sites up to the Fermi level. These charges can be used to calculate the hole densities. The hole carrier density in our calculation and experimentally[7] corresponds to the total number of holes that are introduced in the insulating compound, the ground state of which has the formal electronic configuration $(CuO_2)^{2-}$. Thus one can obtain the hole density from a comparison of the total charge on a CuO_2 plane with the one that will be found in its insulating ground state. The latter has the electronic configuration $(CuO_2)^{2-}$ and thus one obtains a total of 21 valence electrons at a CuO_2 plane, assuming Sr and Ca to be completely ionic Sr^{2+} and Ca^{2+}. As shown in Table I these elements are, however, not fully ionic and retain a small charge at their sites due to a weak hybridization with the neighboring oxygen atoms. The net result of this hybridization is that the total charge on a CuO_2 plane due to Cu and oxygen atoms alone will not be the expected 21 valence electrons in the insulating ground state but somewhat smaller. This difference can be calculated if we assume that all nearest-neighbor oxygen atoms of a given Sr or Ca atom contribute equally to the charge at that site. By comparing the total charge so obtained on a CuO_2 plane with the one given in Table I, the hole densities can be calculated, and are also given in Table I.

Table I shows that the compound Bi-2:2:0:1 is highly overdoped and nearly a metal with a hole density of 0.543 holes/CuO_2. This is much larger than the one 0.283 holes/CuO_2 that we obtained previously[22] for $YBa_2Cu_3O_7$. It has been reported[16] that the T_c in this compound can be raised to ~36 K by the substitution of ~20% La at the Sr site. This should result in a substantial reduction of the hole density in this compound. If we assume that each additional electron brought by the La substitution goes to fill up the hole in the CuO_2 plane, the hole density in the substituted compound would be ~0.143 holes/CuO_2. However, the La substitution also results in an increased uptake of oxygen, which tends to raise the hole density. The latter depends upon the position of the inserted oxygen atoms, and thus it is difficult to make an estimate of the reduction in the hole density brought by La substitution unless the stoichiometry in oxygen can be considered to be unchanged. This large hole density in the Bi-2:2:0:1 compound results from a large electron transfer towards the BiO layers. As one can see from Table I the total charge of ~6.8 electrons on a BiO layer is such that BiO layers are close to being neutral rather than $(BiO)^+$ expected in an ionic picture. Similarly, the valence of Bi in Table I is very close to +1 rather than its formal ionic valence of +3.

The intercalation of a $CaCuO_2$ layer in Bi-2:2:0:1 to form the compound Bi-2:2:1:2 brings no extra charge carriers, since the inserted layer is electrically neutral. The new compound has, however, two CuO_2 planes. If there were no charge redistribution and a simple sharing of the hole densities between the two CuO_2 planes, one would expect a hole density of ~0.271 holes/CuO_2 in Bi-2:2:1:2, which is exactly one half of the value in Bi-2:2:0:1. However, Table I shows that in Bi-2:2:1:2 the hole density is considerably larger ~0.370 holes/CuO_2. This increased hole density arises essentially as a result of an increased electron transfer to the BiO layers. This can be seen in Table I, where one sees that in Bi-2:2:1:2 each BiO layer accommodates an additional 0.092 electron as compared to those in Bi-2:2:0:1. In fact our calculations show that in Bi-2:2:1:2 there is an upward shift of ~0.08

TABLE I. Effective charges at different atomic sites (*p* at oxygen and bismuth sites and *d* at other sites) in $Bi_2Sr_2CuO_6$, $Bi_2Sr_2CaCu_2O_8$, and $Bi_2Sr_2Ca_2Cu_3O_{10}$. The hole counts at CuO_2 planes have been determined as explained in the text.

Site	$Bi_2Sr_2CuO_6$	$Bi_2Sr_2CaCu_2O_8$	$Bi_2Sr_2Ca_2Cu_3O_{10}$
	CuO_2 planes		
Octahedral coordination			
Plane Cu	9.287		
Plane O	5.483		
Total CuO_2 plane	20.254		
Hole count	0.543		
Pyramidal coordination			
Plane Cu		9.353	9.384
Plane O_1		5.491	5.499
Plane O_2		5.490	5.516
Total CuO_2 plane		20.334	20.399
Hole count		0.370	0.309
Square planar coordination			
Plane Cu			9.374
Plane O			5.476
Total CuO_2 plane			20.326
Hole count			0.286
Ca Planes			
Ca		0.402	0.388
SrO Planes			
Sr	0.228	0.214	0.220
O	5.346	5.360	5.367
Total SrO plane	5.574	5.574	5.587
Bi_2O_2 layers			
Bi	1.831	2.004	2.039
O	4.918	4.887	4.924
Total BiO plane	6.799	6.891	6.963
Total charge	45.000	66.000	87.000

eV of the Fermi level, E_F, relative to that in Bi-2:2:0:1. This results in an increased electron transfer to the BiO layers, but also to a lower value of the hole density in each individual CuO_2 plane. The hole density of 0.370 holes/CuO_2 obtained for Bi-2:2:1:2 is far above the value ~0.283 holes/CuO_2 obtained for $YBa_2Cu_3O_7$, which is considered to be near the optimum. A 20% substitution of Y at the Ca site is reported[18] to raise the T_c in this compound from 80 to 90 K. It would thus appear that this substitution brings the hole density to its near optimum value of ~0.28 holes/CuO_2 in this compound.

The trend found in Bi-2:2:1:2 from Bi-2:2:0:1 is also observed in the third compound Bi-2:2:2:3. The presence of an additional $CaCuO_2$ layer results in a further increase in the Fermi energy, E_F by ~0.08 eV and an increased electron transfer to the BiO layers although this is less than what was found in Bi-2:2:1:2 relative to that in Bi-2:2:0:1. As Table I shows the BiO layers in Bi-2:2:2:3 have an excess of 0.072 electron each relative to those in Bi-2:2:1:2. Thus the total hole count per chemical formula is larger in Bi-2:2:2:3 than in Bi-2:2:1:2. On the other hand an upward shift in the Fermi level also results in a lower value of hole density in each CuO_2 plane. Although the hole density in the CuO_2 planes with the pyramidal coordination is larger than in the one with the square planar coordination (0.309 versus 0.286 holes/CuO_2), this difference is not very large. Our calculations thus show that ~32% of the hole density in Bi-2:2:2:3 is localized in the central CuO_2 plane compared to ~34% each in the CuO_2 planes above and below. This is in agreement with the experimental data of Trokiner *et al.*[23] The mean value of the hole density ~0.30 holes/CuO_2 in Bi-2:2:2:3 is quite close to the one obtained in the compound $YBa_2Cu_3O_7$, and appears to be quite optimum. However, considering the fact that $YBa_2Cu_3O_7$ itself is considered very slightly overdoped and the calculated hole density in Bi-2:2:2:3 is already slightly higher than in $YBa_2Cu_3O_7$ it would appear that it should be possible to raise the T_c of Bi-2:2:2:3 through a slight reduction of its hole density.

It is quite customary to present the hole densities in these compounds in terms of the valence of Cu. This is strictly an ionic picture in which Sr and Ca are fully ionic Sr^{2+} and Ca^{2+}, as is also the case with oxygen which is O^{2-}. The valence sharing then occurs between Cu and Bi. Associating the charges at Sr and Ca sites equally with their nearest-neighboring oxygens, we have shown in Table II for the three compounds the total charges at

TABLE II. Total charges at the CuO_2 planes and the Bi_2O_4 layers in $Bi_2Sr_2CuO_6$, $Bi_2Sr_2CaCu_2O_8$, and $Bi_2Sr_2Ca_2Cu_3O_{10}$ obtained by a transfer of the charge at Sr and Ca sites to the neighboring oxygens, as explained in the text, and the effective valence of Cu and Bi obtained from these charges.

Site	$Bi_2Sr_2CuO_6$	$Bi_2Sr_2CaCu_2O_8$	$Bi_2Sr_2Ca_2Cu_3O_{10}$
	CuO_2 planes		
Octahedral coordination			
Total charge	20.457		
Valence Cu	2.543		
Pyramidal coordination			
Total charge		20.630	20.691
Valence Cu		2.370	2.309
Square planar coordination			
Total charge			20.714
Valence Cu			2.286
	Bi_2O_4 layers		
Total charge	24.543	24.740	24.904
Valence Bi	2.729	2.630	2.548

CuO_2 planes, and the Bi_2O_4 layers, which contain oxygens both in the planes of Bi and Sr. In the insulating ground state a Cu atom has the formal ionic configuration Cu^{2+} and thus the CuO_2 plane the formal electronic configuration $(CuO_2)^{2-}$. On the other hand, a Bi atom has a formal ionic configuration Bi^{3+} so that the formal electronic configuration of a Bi_2O_4 layer is $(Bi_2O_4)^{2-}$. With this one would expect 21 valence electrons at a CuO_2 plane and 24 valence electron at a Bi_2O_4 layer. A deficit in charge from these values would mean that the cation donated additional electrons and hence would mean an increased valence, while an increase in charge would indicate a reduced value of valence from the formal value. The valence of Cu and Bi calculated in this manner are shown in Table II. As one can see the valence of Bi is always less than its formal value of 3 and further it decreases in the series Bi-2:2:0:1, Bi-2:2:1:2, and Bi-2:2:2:3. The valence of Cu is also not constant and also decreases in the series but this decrease is much more rapid. If one extrapolates the hole density from Table I or the valence of Cu from Table II to a hypothetical four CuO_2 layer compound, one obtains a mean value ~ 0.24 holes/CuO_2 for the hole density. As for Bi-2:2:2:3 the hole density in the central layers will be even lower, but since the mean value itself is lower than the optimum value, the T_c in this compound is expected to be lower. Such a trend has indeed been observed in the isostructural Tl-based superconductors.[24] These calculations thus indicate that both the number of CuO_2 layers and the hole density in each layer play decisive roles in determining the value of T_c.

IV. CONCLUSIONS

We have shown that the superconductivity in the bismuth cuprate superconductors arises due to the electron transfer from the CuO_2 planes to the BiO_2 layers. The low value of T_c observed in Bi-2:2:0:1 compound can be attributed to its overdoped nature, which makes it a good metal but not a good superconductor. With the increasing number of CuO_2 planes the hole density per CuO_2 plane decreases although the total hole density per chemical formula increases; the latter is due to an increased electron transfer to the BiO_2 layers but this increase is small. The calculated hole density in the two layer compound Bi-2:2:1:2 remains significantly above the optimum value for $YBa_2Cu_3O_7$, while in Bi-2:2:2:3 it is nearly optimum. Our calculations suggest that it might be possible to raise the T_c in this compound still further by a slight reduction in the hole density. Further, we expect that the T_c in a hypothetical four-layer material will be lower than in Bi-2:2:2:3.

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