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## Theoretical study of the chemical pressure effect on $T_c$ in the cuprate superconductors

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### Abstract

Application of physical pressure on the cuprate superconductors often results in an enhancement of  $T_c$ . Motivated by this fact, we study the *chemical pressure* effect on the single-layered  $\text{La}_2\text{CuO}_4$  and  $\text{HgBa}_2\text{CuO}_4$  starting from the two-orbital Hubbard model deduced from a first-principles calculation. It is shown that the chemical pressure effects induced by La-site substitution in  $\text{La}_2\text{CuO}_4$  or Hg-site substitution in  $\text{HgBa}_2\text{CuO}_4$  are not expected to be effective for raising  $T_c$ .

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**Keywords:** cuprates; superconductivity; two-orbital model; band calculation; fluctuation exchange approximation (FLEX); chemical pressure

### 1. Introduction

One of the interesting features of the high  $T_c$  cuprates is its  $T_c$  variance against physical pressure application[1]. In a number of cases,  $T_c$  is enhanced by applying hydrostatic pressure[2-5], and understanding the origin of this phenomenon should be important in obtaining a guiding principle toward synthesizing higher  $T_c$  materials. Recently, we have theoretically analysed the effect of the pressure using a two orbital model[5,6] consisting of the  $3d_{x^2-y^2}$  and the  $3d_{z^2}$ [7-19] orbitals, and pinned down three  $T_c$ -controlling factors that are affected by pressure[20], namely, (i) the level off-set  $\Delta E$  between the two orbitals, (ii) the warping of the Fermi surface that is affected by Cu(4s) hybridization[21,22], and (iii) the band width. Since the physical pressure has significant effects on  $T_c$ , one may

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wonder whether it is possible to obtain a similar effect through element substitution, i.e., the chemical pressure.

In the present work, we investigate the chemical pressure effect on two typical single-layered cuprates,  $\text{La}_2\text{CuO}_4$  and  $\text{HgBa}_2\text{CuO}_4$ . In doing so, we consider hypothetical materials  $X_2\text{CuO}_4$  ( $X=\text{Sc}, \text{Y}, \text{La}$ ) and  $X\text{Ba}_2\text{CuO}_4$  ( $X=\text{Zn}, \text{Cd}, \text{Hg}$ ), and estimate the effects of the chemical pressure effect induced by the element substitution on the superconductivity. Our ultimate goal along this line of study is to consider the possibility of new cuprates which would have higher  $T_c$  than those which are already discovered.

## 2. Calculation methods

We first perform first-principles band calculation to determine the structural parameter of the materials mentioned above. We use VASP code[23] to optimize the total energy of these materials. The lattice constants obtained are  $a=3.82\text{\AA}$  and  $c=13.2\text{\AA}$  for  $\text{La}_2\text{CuO}_4$ ,  $a=3.94\text{\AA}$  and  $c=9.84\text{\AA}$  for  $\text{Hg}_2\text{CuO}_4$ . As for the existing materials, the theoretical estimation agrees well with the experimental results[24]. Using the obtained structures, we construct maximally localized Wannier orbitals [25] to obtain the hopping integrals for the present two-orbital model, in which we consider the  $3d_{x^2-y^2}$  and the  $3d_{z^2}$  orbitals explicitly.

### 2.1. Applying the fluctuation exchange approximation

The electron-electron interactions considered in the present study are the following: the on-site intra-orbital Coulomb repulsion  $U$ , the inter-orbital repulsion  $U'$ , the Hund's coupling  $J$  and pair-hopping  $J'$ . Here we observe the orbital rotational symmetry,  $U-U'=2J$ . We fix the values at  $U=3.0\text{ eV}$ ,  $U'=2.4\text{ eV}$  and  $J=J'=0.3\text{ eV}$ . In recent estimations with first-principles,  $U$  in the cuprates is considered to be  $7-10t$  (namely, about  $3-4.5\text{ eV}$  with  $t$  being the transfer energy) and  $J(J')\sim 0.1U$ , so the values chosen here are within the widely accepted range.

Then we apply the fluctuation exchange approximation (FLEX)[26-28] to the present model to obtain the Green's function renormalized by the many-body self-energy correction. In FLEX, the contributions from bubble and ladder diagrams are included in the self-energy, for which we solve the Dyson's equation in a self-consistent manner. Then we substitute the Green's function to the linearized Eliashberg equation to evaluate the strength of the superconducting instability. The eigenvalue  $\lambda$  of the Eliashberg equation reaches unity at  $T_c$ , so we can use  $\lambda$  at a fixed temperature as a measure of  $T_c$ . We set  $T=0.02\text{ eV}$ , and the number of electrons per copper site to be  $n=2.85$  (i.e., 15% doped in the main band). We take the  $32\times 32\times 4$   $k$ -point meshes and 1024 Matsubara frequencies.

## 3. Results and discussion

### 3.1. Structural deformation

Fig.1 shows the obtained lattice constants of  $X_2\text{CuO}_4$  and  $X\text{Ba}_2\text{CuO}_4$ . As for  $X_2\text{CuO}_4$ , the lattice constants  $a$  and  $c$  decrease with the substitution due to the reduction in the ionic radius. Here, note that  $a$  is similar between the Y- and Sc- compounds, while  $c$  is significantly different. By contrast, for  $X\text{Ba}_2\text{CuO}_4$  the chemical pressure effect on the lattice constants is relatively small. This may reflect the fact that there is more room around the Hg-site in  $X\text{Ba}_2\text{CuO}_4$  than around the La-site in  $X_2\text{CuO}_4$ .

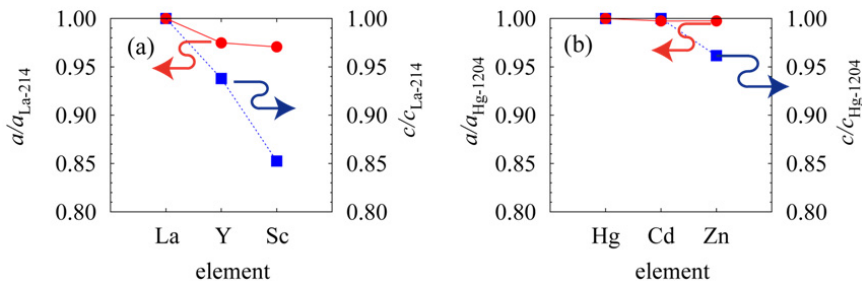


Fig. 1. Obtained lattice constant  $a$  (red circle) and  $c$  (blue square) for (a)  $X_2\text{CuO}_4$  ( $X=\text{Sc}, \text{Y}, \text{La}$ ) (b)  $X\text{Ba}_2\text{CuO}_4$  ( $X=\text{Zn}, \text{Cd}, \text{Hg}$ ).

### 3.2. Chemical pressure effect on $T_c$

Now, we come to the superconductivity. Figure 2 shows the eigenvalue  $\lambda$  of the Eliashberg equation. As for  $X_2\text{CuO}_4$ , while the Y-substitution gives rise to a small enhancement in  $\lambda$ , the Sc-substitution suppresses  $\lambda$ . In  $X_2\text{CuO}_4$ , the  $d_{z^2}$ -orbital-mixture mainly controls the pressure effect on  $T_c$  as revealed in [20], so we plot the element dependence of  $\Delta E$  in panel (a-2). A slight increase in  $\Delta E$  in the Y-compound is due to a reduction of the Cu-O bond length (from a larger crystal field), while the reduction of  $\Delta E$  in the Sc-compound originates from a large reduction of the aspect ratio  $h_0/l$  (Fig.2(a-1), with  $l$  being the in-plane Cu-O bond length) dominating over the Cu-O length reduction effect. As expected,  $\lambda$  and  $\Delta E$  vary in a correlated manner. As for  $X\text{Ba}_2\text{CuO}_4$ , reflecting the small structural change by element substitution,  $\lambda$  changes only slightly as shown in Fig.2(b). In total, these results show that the chemical pressure effect induced by substituting the La-site in  $\text{La}_2\text{CuO}_4$  or the Hg-site in  $\text{HgBa}_2\text{CuO}_4$  is not effective for raising the  $T_c$ .

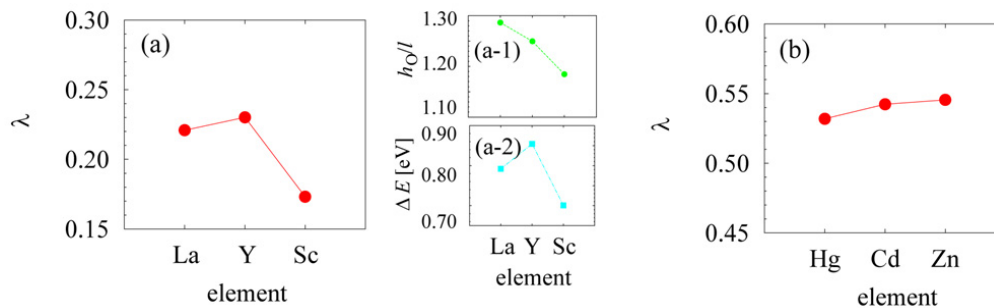


Fig. 2. The eigenvalue  $\lambda$  of Eliashberg equation for (a)  $X_2\text{CuO}_4$  ( $X=\text{Sc}, \text{Y}, \text{La}$ ) (b)  $X\text{Ba}_2\text{CuO}_4$  ( $X=\text{Zn}, \text{Cd}, \text{Hg}$ ). The panels (a-1) and (a-2) show the element dependence of structural and electronic parameters (see text).

### 4. Conclusion

To summarize, we have studied the chemical pressure effect on  $\text{La}_2\text{CuO}_4$  and  $\text{HgBa}_2\text{CuO}_4$  with first-principles calculations. For hypothetical compounds,  $\text{Sc}_2\text{CuO}_4$ ,  $\text{Y}_2\text{CuO}_4$ ,  $\text{CdBa}_2\text{CuO}_4$  and  $\text{ZnBa}_2\text{CuO}_4$ , we have estimated the lattice parameter with a first-principles optimization. Applying the FLEX approximation to the two-orbital Hubbard model constructed from the maximally localized Wannier orbitals for each compound, we have revealed that the chemical pressure due to these substitution is not effective in raising  $T_c$ .

This result leads us to expect that some other kind of chemical pressure may enhance  $T_c$ , since we know that applying physical pressure to  $\text{HgBa}_2\text{CuO}_4$  results in a large enhancement of  $T_c$ . In this sense, the Ba-site substitution in  $\text{HgBa}_2\text{CuO}_4$  can in fact have entirely different effects, which will be published elsewhere.

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