

Bound hole states and ferrons in $\text{La}_2\text{CuO}_{4+\delta}$

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Bound hole states induced by excess oxygen in $\text{La}_2\text{CuO}_{4+\delta}$ are studied in the framework of the extended Hubbard model, using the spin-wave approximation and the Lanczos algorithm. We interpret the band at 0.5 eV in the impurity reflectivity spectrum as connected with the transition from the lowest bound hole level ($S_z=1/2$) to the unperturbed hole band. It is shown that in the realistic parameter range the excess O^{2-} ion does not lead to the formation of ferrons. [S0163-1829(98)03818-1]

At present it is well established that structural defects in La_2CuO_4 (rare-earth substitutions, excess oxygen ions) lead to the rise of the carrier (hole) concentration in CuO_2 planes. Along with the creation of free carriers these defects induce bound states in the hole energy spectrum of CuO_2 planes. These states were detected in different experiments. Of special interest are measurements of the reflectivity spectrum of the antiferromagnetically ordered $\text{La}_2\text{CuO}_{4+\delta}$,¹ where two bands (0.13 and 0.5 eV) in the forbidden gap were observed. In Ref. 2 we considered the respective bound states on the supposition that they are connected with the lowest bands characterized by the z component of the total spin $S_z = \pm 1/2$. We used approximate band states obtained in Ref. 3.

Some experiments, e.g., EPR data in Ref. 4, indicate the formation of small ferromagnetically ordered regions around holes on the antiferromagnetic background of CuO_2 planes. Such elementary excitations called ferrons correspond to $|S_z| > 1/2$. Calculations³ based on the t - J model showed that in the periodic lattice the ferrons are energetically unfavorable for parameters of cuprate perovskites. However, these crystals always contain defects—oxygen vacancies, interstitial, and substitutional impurities which have a pronounced effect on the properties of lightly doped samples used in the mentioned experiments. These defects can be supposed to stabilize the ferrons.

In the present paper we investigate this process for the interstitial O^{2-} ion. We use the extended Hubbard model and the Lanczos algorithm to obtain the lowest eigenstates with different S_z . We have found that the interstitial O^{2-} ion does not lead to the ferron formation, as the ground state is characterized by $S_z = \pm 1/2$ for parameters of cuprate perovskites. The obtained results confirm the conclusion of Ref. 2 that the spectral band at 0.5 eV in the reflectivity spectrum¹ of $\text{La}_2\text{CuO}_{4+\delta}$ is connected with the transitions from this bound ground state to the band of extended hole states.

The extended Hubbard model is used for the description of the electronic states in a CuO_2 plane. The Hamiltonian can be written in the form⁵

$$H_0 = \sum_{\mathbf{m}} H_{\mathbf{m}} + 2t\lambda_{\mathbf{a}} \sum_{\mathbf{m}\sigma} (d_{\mathbf{m}\sigma}^\dagger \phi_{\mathbf{m}+\mathbf{a},\sigma} + \text{H.c.}),$$

$$H_{\mathbf{m}} = U n_{\mathbf{m},+1} n_{\mathbf{m},-1} + \Delta \sum_{\sigma} \phi_{\mathbf{m}\sigma}^\dagger \phi_{\mathbf{m}\sigma} + 2t\lambda_0 \sum_{\sigma} (d_{\mathbf{m}\sigma}^\dagger \phi_{\mathbf{m}\sigma} + \text{H.c.}), \quad (1)$$

where $d_{\mathbf{m}\sigma}^\dagger$ is the creation operator of electrons in the $3d_{x^2-y^2}$ orbitals of copper at the plane site \mathbf{m} with the spin $\sigma = \pm 1$, $\phi_{\mathbf{m}\sigma}^\dagger$ is the Fourier transform of the operator $\phi_{\mathbf{k}\sigma}^\dagger = (\beta_{\mathbf{k}}/2\sqrt{N}) \sum_{\mathbf{m}\mathbf{a}} \exp(-i\mathbf{k}\mathbf{m}) p_{\mathbf{m}+\mathbf{a}/2,\sigma}^\dagger$ constructed from the creation operators $p_{\mathbf{m}+\mathbf{a}/2,\sigma}^\dagger$ of electrons in the $2p_\sigma$ orbitals of oxygen. Complementary linear combinations of these operators, which do not hybridize with the $3d_{x^2-y^2}$ copper orbitals are omitted in Eq. (1) because their energy levels are much higher in energy. $\mathbf{a} = (\pm a, 0), (0, \pm a)$, where a is the in-plane copper distance, $\beta_{\mathbf{k}} = \{1 + [\cos(k_x a) + \cos(k_y a)]/2\}^{-1/2}$, N is the number of sites; U , Δ , and t are the Hubbard repulsion on copper, the Cu-O promotion energy and hybridization, respectively, $n_{\mathbf{m}\sigma} = d_{\mathbf{m}\sigma}^\dagger d_{\mathbf{m}\sigma}$, $\lambda_{\mathbf{m}} = N^{-1} \sum_{\mathbf{k}} \exp(i\mathbf{k}\mathbf{m}) \beta_{\mathbf{k}}^{-1}$, $\lambda_0 \approx 0.96$, $\lambda_{\mathbf{a}} \approx 0.14$; the other components of $\lambda_{\mathbf{m}}$ are small and the respective terms are omitted in Eq. (1).

An interstitial oxygen ion O^{2-} is positioned above a plane oxygen halfway between two neighboring CuO_2 planes.⁶ To be specific we suppose that the ion is located above the plane oxygen site $\mathbf{l} - \mathbf{y}/2$, and add the following terms to the Hamiltonian:

$$H_1 = V_{\text{Cu}}(n_1 + n_{1-\mathbf{y}}) + V_{\text{O}} n_{1-\mathbf{y}/2}. \quad (2)$$

Here $n_1 = \sum_{\sigma} n_{1\sigma}$, $n_{1-\mathbf{y}/2} = \sum_{\sigma} p_{1-\mathbf{y}/2,\sigma}^\dagger p_{1-\mathbf{y}/2,\sigma}$, $\mathbf{y} = (0, a)$. By analogy with the Wannier exciton⁷ the dielectric description with the high-frequency dielectric constant ϵ_∞ can be used for potentials V_{Cu} and V_{O} as the distances r_{Cu} and r_{O} between the interstitial ion and the respective plane sites are

larger than $(\hbar^2/mE_G)^{1/2} \leq 1 \text{ \AA}$, where $m \sim 10 m_0$ is the carrier effective mass⁸ and $E_G \approx 2 \text{ eV}$ is the forbidden gap.⁹ Thus,

$$V_{\text{Cu,O}} = \frac{2e^2}{\epsilon \infty r_{\text{Cu,O}}} \quad (3)$$

[we use the electron picture in Eq. (2), therefore potentials V_{Cu} and V_{O} are positive]. In notations of Eq. (1) terms (2) can be rewritten as

$$H_i = \sum_{\sigma} V_{\text{O}} \frac{(\beta_0 + \beta_y)^2}{4} (\phi_{1\sigma}^{\dagger} \phi_{1\sigma} + \phi_{1-y,\sigma}^{\dagger} \phi_{1-y,\sigma} + \phi_{1\sigma}^{\dagger} \phi_{1-y,\sigma} + \phi_{1-y,\sigma}^{\dagger} \phi_{1\sigma}) + V_{\text{Cu}}(n_{1+} + n_{1-y}), \quad (4)$$

where β_0 and β_y are the components of the Fourier transform of $\beta_{\mathbf{k}}$ for $\mathbf{m} = \mathbf{0}$ and $\mathbf{m} = \mathbf{y}$, respectively. $(\beta_0 + \beta_y)^2 \approx 0.919$, the other components are small.

For the low-energy part of the spectrum and for parameters¹⁰ of La_2CuO_4 Hamiltonian (1) can be reduced⁵ to the effective t - J Hamiltonian. This reduction is based on the separation of the Hamiltonian into the one- and two-site parts, as given in Eq. (1). These two parts are characterized by energy parameters differing by one order of magnitude, which provides an appropriate starting point for the perturbation theory. The zero-order, one-site, part $\sum_{\mathbf{m}} H_{\mathbf{m}}$ of the Hamiltonian has two sets of states well-separated from other states. These states, which correspond to unoccupied and occupied states of the t - J model, can be written in the form

$$|\mathbf{m} 2\rangle = \left[c_{21} \frac{1}{\sqrt{2}} (\phi_{\mathbf{m},+1}^{\dagger} d_{\mathbf{m},-1}^{\dagger} - \phi_{\mathbf{m},-1}^{\dagger} d_{\mathbf{m},+1}^{\dagger}) + c_{22} \phi_{\mathbf{m},-1}^{\dagger} \phi_{\mathbf{m},+1}^{\dagger} + c_{23} d_{\mathbf{m},-1}^{\dagger} d_{\mathbf{m},+1}^{\dagger} \right] |v_{\mathbf{m}}\rangle,$$

$$|\mathbf{m} 3\sigma\rangle = [c_{31} \phi_{\mathbf{m},+1}^{\dagger} \phi_{\mathbf{m},-1}^{\dagger} d_{\mathbf{m}\sigma}^{\dagger} + c_{32} \phi_{\mathbf{m}\sigma}^{\dagger} d_{\mathbf{m},-1}^{\dagger} d_{\mathbf{m},+1}^{\dagger}] |v_{\mathbf{m}}\rangle, \quad (5)$$

where $|v_{\mathbf{m}}\rangle$ is the site vacuum state and the coefficients c_{ij} are obtained in the course of the diagonalization of $H_{\mathbf{m}}$. Omitting unessential terms, Hamiltonians (1) and (4) can be rewritten in terms of the state vectors (5) as

$$H_0 = t_{\text{eff}} \sum_{\mathbf{m}\mathbf{a}\sigma} |\mathbf{m} + \mathbf{a}, 3\sigma\rangle \langle \mathbf{m} + \mathbf{a}, 2| |\mathbf{m} 2\rangle \langle \mathbf{m} 3\sigma| + \frac{J}{2} \sum_{\mathbf{m}\mathbf{a}} \mathbf{S}_{\mathbf{m}} \cdot \mathbf{S}_{\mathbf{m}+\mathbf{a}}, \quad (6)$$

$$H_i = \sum_{\mathbf{i}'} \epsilon |\mathbf{i}' 2\rangle \langle \mathbf{i} 2| + t_1 \sum_{\sigma} (|\mathbf{1} 2\rangle |\mathbf{1-y} 3\sigma\rangle \langle \mathbf{1-y} 2| \langle \mathbf{1} 3\sigma| + \text{H.c.}), \quad (7)$$

where

$$S_{\mathbf{m}}^{\sigma} = S_{\mathbf{m}}^x + i\sigma S_{\mathbf{m}}^y = |\mathbf{m} 3\sigma\rangle \langle \mathbf{m} 3, -\sigma|,$$

$$S_{\mathbf{m}}^z = \frac{1}{2} \sum_{\sigma} \sigma |\mathbf{m} 3\sigma\rangle \langle \mathbf{m} 3\sigma|,$$

are the components of the spin operator $\mathbf{S}_{\mathbf{m}}$,

$$\epsilon = \frac{V_{\text{O}}(\beta_0 + \beta_y)^2}{4} (2c_{22}^2 + c_{21}^2 - 2c_{31}^2 - c_{32}^2) + V_{\text{Cu}}(c_{21}^2 + 2c_{23}^2 - c_{31}^2 - 2c_{32}^2),$$

$$t_1 = \frac{V_{\text{O}}(\beta_0 + \beta_y)^2}{4} \left(\frac{c_{31}c_{21}}{\sqrt{2}} - c_{23}c_{32} \right)^2,$$

$\mathbf{i}' = \mathbf{i}, \mathbf{i-y}$, and t_{eff} and J are the effective hopping and superexchange constants, respectively (in terms of the coefficients c_{ij} and site energies these constants are given in Ref. 5).

Further simplifications of Hamiltonians (6) and (7) can be achieved with the use of the spin-wave approximation^{3,11} which has been shown to be remarkably accurate in the description of undoped and lightly doped samples where the number of magnons is small.^{3,12} The approximation reduces to neglecting terms of the third and higher orders in the spin-wave operators $b_{\mathbf{m}}$ introduced into Eq. (6) by

$$S_{\mathbf{m}}^{+1} = \Phi_{\mathbf{m}} b_{\mathbf{m}} P_{\mathbf{m}}^{+1} + b_{\mathbf{m}}^{\dagger} \Phi_{\mathbf{m}} P_{\mathbf{m}}^{-1}, \quad S_{\mathbf{m}}^{-1} = (S_{\mathbf{m}}^{+1})^{\dagger},$$

$$S_{\mathbf{m}}^z = e^{i\text{Im}} \left(\frac{n_{\mathbf{m}}}{2} - b_{\mathbf{m}}^{\dagger} b_{\mathbf{m}} \right),$$

where $\Phi_{\mathbf{m}} = (1 - b_{\mathbf{m}}^{\dagger} b_{\mathbf{m}})^{1/2}$, $\mathbf{\Pi} = (\pi/a, \pi/a)$, $P_{\mathbf{m}}^{\sigma} = [1 + \sigma \exp(i\mathbf{\Pi}\mathbf{m})]/2$.

Let us introduce the hole creation operator $h_{\mathbf{m}}^{\dagger} = \sum_{\sigma} P_{\mathbf{m}}^{\sigma} |\mathbf{m} 2\rangle \langle \mathbf{m} 3\sigma|$ for the Néel state $|\mathcal{N}\rangle = \mathbf{\Pi}_{\mathbf{m}} (\sum_{\sigma} P_{\mathbf{m}}^{\sigma} |\mathbf{m} 3\sigma\rangle)$ (the second Néel state and the respective hole operator can be obtained by substituting $P_{\mathbf{m}}^{\sigma}$ with $1 - P_{\mathbf{m}}^{\sigma}$ in these formulas). After the diagonalization of the Heisenberg part of the Hamiltonian, it reads

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_i = \sum_{\mathbf{m}\mathbf{m}'\mathbf{a}} t_{\text{eff}} [h_{\mathbf{m}+\mathbf{a}} h_{\mathbf{m}}^{\dagger} b_{\mathbf{m}-\mathbf{m}'}^{\dagger} (u_{\mathbf{m}'+\mathbf{a}} + v_{\mathbf{m}'}) + \text{H.c.}] + \frac{J}{2} \sum_{\mathbf{m}\mathbf{m}'} \omega_{\mathbf{m}'} b_{\mathbf{m}}^{\dagger} b_{\mathbf{m}+\mathbf{m}'}$$

$$+ \epsilon (\tilde{n}_1 + \tilde{n}_{1-y}) + t_1 \sum_{\mathbf{m}\mathbf{m}'\mathbf{a}} [h_{\mathbf{m}+\mathbf{a}} h_{\mathbf{m}}^{\dagger} b_{\mathbf{m}-\mathbf{m}'}^{\dagger} [\delta_{\mathbf{l},\mathbf{m}} \delta_{\mathbf{a},-y} + \delta_{\mathbf{l},-\mathbf{y},\mathbf{m}} \delta_{\mathbf{a},y}] (u_{\mathbf{m}'+\mathbf{a}} + v_{\mathbf{m}'}) + \text{H.c.}], \quad (8)$$

where $u_{\mathbf{m}}$, $v_{\mathbf{m}}$ and $\omega_{\mathbf{m}}$ are the Fourier transforms of $\cosh(\alpha_{\mathbf{k}})$, $-\sinh(\alpha_{\mathbf{k}})$, and $\omega_{\mathbf{k}}=4\sqrt{1-\gamma_{\mathbf{k}}^2}$, respectively, $\alpha_{\mathbf{k}}=(1/8)\ln[(1+\gamma_{\mathbf{k}})/(1-\gamma_{\mathbf{k}})]$, $\gamma_{\mathbf{k}}=[\cos(k_x a)+\cos(k_y a)]/2$, $\tilde{n}_1=h_1^\dagger h_1$. Due to the fast decrease of $u_{\mathbf{m}'+\mathbf{a}}+v_{\mathbf{m}'}$ and $\omega_{\mathbf{m}'}$ with the growth of $|\mathbf{m}'|$ only the components with $\mathbf{m}'=(\pm a,0),(0,\pm a)$ for the sum in the first kinetic energy term and the components with $\mathbf{m}'=(0,0),(\pm a,\pm a),(\pm 2a,0),(0,\pm 2a)$ for $\omega_{\mathbf{m}'}$ in the second magnetic energy term need to be retained in Eq. (8) in the subsequent calculations.

To investigate the lowest eigenvalues and eigenfunctions of Hamiltonian (8) we use the modified Lanczos algorithm.^{3,13} In one step of this algorithm we determine a final state $|\mathbf{f}\rangle$ from an initial state $|\mathbf{i}\rangle$ according to the procedure

$$\langle \mathbf{i}|\mathbf{i}\rangle=1, \mathcal{E}_i=\langle \mathbf{i}|\mathcal{H}|\mathbf{i}\rangle, \quad \mathcal{V}|\mathbf{f}\rangle=(\mathcal{H}-\mathcal{E}_i)|\mathbf{i}\rangle. \quad (9)$$

Here \mathcal{V} is the normalization constant which is determined from the condition $\langle \mathbf{f}|\mathbf{f}\rangle=1$. After one Lanczos step the energy is minimized in the subspace of the states $|\mathbf{i}\rangle$ and $|\mathbf{f}\rangle$ [it follows from (9) that $\langle \mathbf{i}|\mathbf{f}\rangle=0$]. The obtained new initial function $|\mathbf{i}'\rangle=c_1|\mathbf{i}\rangle+c_2|\mathbf{f}\rangle$ is used in the next Lanczos step. This procedure is continued until the necessary accuracy of the eigenvalue is reached. If the initial function $|\mathbf{i}\rangle$ is characterized by some spin projection S_z , $|\mathbf{f}\rangle$ also corresponds to the same S_z . Therefore, it is possible to obtain the lowest eigenvalues of Hamiltonian (8) with a given S_z by starting from a state with the respective S_z . Notice that states with $S_z>1/2$ contain flipped spins near the hole and correspond to ferrons.

The number of spin configurations necessary for describing the states $|\mathbf{i}\rangle$ and $|\mathbf{f}\rangle$ grows from step to step. To avoid an overflow of the computer storage and to reduce the growth of the computation time, by analogy with Ref. 3 in each step the state $|\mathbf{i}'\rangle$ is restricted to the j configurations with the largest amplitudes. After normalization this restricted state is used as the initial state for the next recursion step of the Lanczos procedure (9). We used $j=200$. The $|\mathbf{f}\rangle$ state obtained from such $|\mathbf{i}'\rangle$ state and used for the calculation of the eigenvalue contains of the order of 10^4 spin configurations. This is comparable to the number of configurations used in exact diagonalization studies of small lattices. To check the accuracy of the obtained results in some cases we increased j up to 250 which led only to small quantitative changes (of the order of $10^{-4}t_{\text{eff}}$) in the eigenvalue.

With the modified Lanczos algorithm outlined above we have obtained the lowest localized hole states induced by the excess oxygen in $\text{La}_2\text{CuO}_{4+\delta}$ and characterized by $S_z \leq 13/2$. For parameters of this crystal the ratio J/t_{eff} can be estimated to lie in the range 0.2–0.5.¹⁰ For $J/t_{\text{eff}}=0.42$ the calculated energies are listed in Table I. The high-frequency dielectric constant¹ $\epsilon_\infty=5$ was used in these calculations.

One can see that the lowest eigenvalues E_B of Hamiltonian (8) increase monotonically with S_z . This result is a consequence of the energy position of hole bands with different S_z . As known,^{3,14–17} for the used parameters the lowest hole band corresponds to $S_z=1/2$. For the considered two-dimensional system bound states are split off from the band already at infinitely small perturbation for the major part of symmetries. This means that at least for small pertur-

TABLE I. Bound energy levels E_B for different S_z at $J/t_{\text{eff}}=0.42$ and $\epsilon_\infty=5$. E_0 is the center of gravity of the band.

S_z	Energy, eV		
	E_0	E_B	E_0-E_B
1/2	-0.454	-1.029	0.575
3/2	-0.343	-0.915	0.572
5/2	-0.218	-0.751	0.533

bations relative positions of the bound states with different S_z are the same as the positions of the corresponding bands and the bound state with $S_z=1/2$ is energetically most favorable. To estimate the splitting of the bound hole states from the unperturbed hole bands with the same S_z we have calculated the energies of the localized hole states E_0 in the case of zero perturbation $\epsilon=t_1=0$. These energies correspond to the center of gravity of the respective bands and, according to Ref. 14, are close to the bottoms of the bands. Table I shows that $E_0-E_B \approx 0.5$ eV. This energy difference is close to the value found in Ref. 2 with approximate wave functions of the band states obtained in the first step of the described Lanczos algorithm. This confirms the conclusion made in Ref. 2 that the maximum at 0.5 eV in the reflectivity spectra¹ of $\text{La}_2\text{CuO}_{4+\delta}$ is connected with transitions from the lowest bound state $S_z=1/2$ to the band states with the same S_z . This process can be considered as the optical ionization of the bound state. The second band, at 0.13 eV, observed in the spectra and ascribed in Ref. 2 to the transition from the excited bound state to the band states, cannot be considered in the used version of the Lanczos algorithm which gives the lowest bound state only.

To investigate the effect of the interstitial O^{2-} ion on the ferron formation we have calculated the lowest eigenvalues of Hamiltonian (8) with $S_z \leq 13/2$. As seen in Fig. 1, S_z of the ground state grows with decreasing J . The same behavior has been obtained for band hole states^{3,14,15} and corresponds to the growth of the ferron size with decreasing J . Such

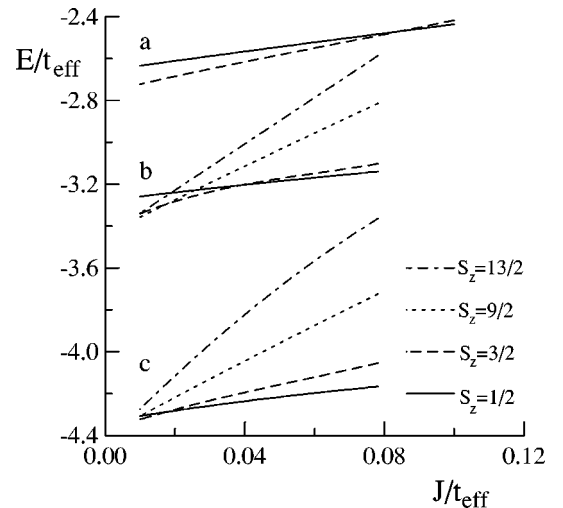


FIG. 1. The dependence of energies of the localized hole states with $S_z=1/2, 3/2, 9/2,$ and $13/2$ on J/t_{eff} for different perturbations induced by the defect. (a) $\epsilon=t_1=0$; (b) $\epsilon=-t_{\text{eff}}, t_1=0.3t_{\text{eff}}$; (c) $\epsilon=-2t_{\text{eff}}, t_1=0.6t_{\text{eff}}$.

dependences of eigenvalues on the ratio J/t are also in satisfactory agreement with the results of the exact diagonalization on small clusters.^{16,17} Notice, however, that the first change of S_z in the ground state from 1/2 to 3/2 takes place at $J/t_{\text{eff}} \approx 0.08$ when $\epsilon = t_1 = 0$. For larger S_z and with increasing the perturbation induced by the defect the respective ratio J/t_{eff} becomes even smaller. As mentioned above, for parameters of cuprate perovskites J/t_{eff} is estimated to lie in the range 0.2–0.5.¹⁰ Thus the interstitial O^{2-} defect does not promote the formation of ferrons.

In summary, with the use of the spin-wave approximation and the Lanczos algorithm the bound hole states induced by interstitial O^{2-} ions in $\text{La}_2\text{CuO}_{4+\delta}$ were investigated. For parameters of $\text{La}_2\text{CuO}_{4+\delta}$ the lowest bound state is charac-

terized by $S_z = 1/2$ in a wide range of defect potentials. This means that the O^{2-} defect does not promote the formation of ferrons. The obtained results allow us to identify the 0.5 eV band observed in the reflectivity spectrum of this crystal as connected with the transition (optical ionization) from the defect ground state with $S_z = 1/2$ to the band of extended states with the same S_z .

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