Proper reduction scheme to an extended *t-J* model and the hole dispersion in $Sr_2CuO_2Cl_2$

V. Yu. Yushankhai and V. S. Oudovenko
Joint Institute for Nuclear Research, 141980 Dubna, Moscow Region, Russian Federation

R. Hayn
Max Planck Arbeitsgruppe Elektronensysteme, Technische Universität Dresden, D-01062 Dresden, Federal Republic of Germany

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The reduction of the three-band *p-d* model for the CuO_2 plane in cuprates with the standard set of parameters as derived by Hybertsen *et al.* [Phys. Rev. B **41**, 11 068 (1990)] to an effective t-t'-t''-J model with a three-site hopping term is discussed in detail in the framework of the cell-perturbation method. The reduction procedure is formulated to avoid any ambiguity in obtaining the correct signs of the hopping parameters in the effective model, and the contributions of first and second order resulting from the intercell hopping are distinguished. Based on the so-defined extended *t-J* model, the dispersion of a single hole in an antiferromagnetic background is calculated both by a variational ansatz and within the self-consistent Born approximation (SCBA). An extension of the variational method allows us to investigate the influence of a three-site hopping term both on the mean-field level and beyond this approximation. The latter corrections are shown to be small. Both methods give the coinciding result that the additional hopping terms lead to a more isotropic minimum of the dispersion at the point $(\pi/2,\pi/2)$ in k space and to a slightly increased bandwidth in comparison to the pure *t-J* model. These results are compared with recent photoemission measurements for $Sr_2CuO_2Cl_2$ [Wells *et al.*, Phys. Rev. Lett. **74**, 964 (1995)], showing an improvement against the pure *t-J* model, but no complete agreement. In particular, a small anisotropy in the calculated effective masses remains. The lifetime of the hole quasiparticles is investigated within the SCBA, and it is shown that the upper part of the hole spectrum loses its quasiparticle character due to a strong increase of damping if the bare hole bandwidth exceeds a certain threshold value of roughly $2J$. [S0163-1829(97)02623-4]

I. INTRODUCTION

Recent angle-resolved photoemission spectroscopy (ARPES) measurements[1] of the insulating, antiferromagnetic compound $Sr_2CuO_2Cl_2$ provided a good test for several theories of the electronic structure of cuprate superconductors. For the first time it was possible to measure a valence-band dispersion in an undoped compound, which corresponds to the well-defined theoretical problem of the one-hole motion in an antiferromagnetic background. The position of the dispersion minimum at the point $(\pi/2,\pi/2)$ (in the notation of the two-dimensional square lattice) and the measured bandwidth, which is roughly twice the value of the exchange integral J, are in good agreement with predictions of theories based on the pure *t-J* model.[2–6] However, large deviations from these predictions have been found in the shape of the dispersion, especially along the line $(0,0)$-$(\pi,0)$. Also in contrast to the *t-J* model, the experiment[1] indicated that the dispersion near the minimum is highly isotropic. Subsequent theoretical investigations[7–9] have shown that better agreement with the experiment could be obtained if one extends the *t-J* model by taking into account a second hopping integral t_2 (or t' in other notation) to next nearest neighbors[7] and the so-called three-site hopping term.[8,9] Up to now, the additional hopping parameters are treated usually as phenomenological ones. So a further justification both for the form of the extended *t-J* model and for the numerical values of its parameters is strongly required from first-principle calculations. That is especially important in the light of more recent ARPES measurements by Schmidt *et al.*,[10] who confirmed in main features the results by Wells *et al.* for $Sr_2CuO_2Cl_2$, but revealed a noticeable dispersion anisotropy near the minimum.

Usually, first-principle calculations are understood as to start with the determination of the band structure within the local-density approximation (LDA). These LDA calculations, however, have severe difficulties in describing the electronic structure of $Sr_2CuO_2Cl_2$ or similar compounds such as La_2CuO_4, for instance.[11,12] In this approach a metallic behavior is predicted for the undoped compound where the bandwidth and the shape of the band crossing the Fermi level differ substantially from the ones obtained in the ARPES experiment.[1] The reason consists in the insufficient treatment of the strong electron correlations in the LDA calculations. So the question arises if there is any chance to calculate the one-hole dispersion in $Sr_2CuO_2Cl_2$ in a ''first-principle'' sense. A possible solution for that problem can be proposed in two steps. As the first step *A* one has to construct an effective many-band Hubbard-like Hamiltonian (such as the *p-d* model) whose parameters are determined by a constrained density-functional calculation. That has been carried through, for instance, for La_2CuO_4.[13] Band-structure calculations for $Sr_2CuO_2Cl_2$ show only few differences: the additional Cl atoms influence only the states far away from the Fermi level.[11] Therefore, we will assume here that the parameters for La_2CuO_4 are representative for $Sr_2CuO_2Cl_2$ as well. The second step *B* deals with the dispersion of one hole in the valence band. Since we are interested in the low-energy electronic excitations, the high-lying orbitals should be eliminated. So it is reasonable first to reduce the derived

multiband model to an effective t-J-like Hamiltonian and then to calculate the dispersion for this one-band Hamilton operator which is restricted to the low-energy physics only.

In the present paper we adopt the solution for step A as it was presented in Ref. 13 and elaborate step B in great detail and with all the necessary accuracy. We will address two points, namely, the reduction of the three-band p-d model to an effective t-J-like Hamiltonian and the calculation of the one-hole motion characteristics in this extended t-J model. There exist already publications that deal with one part of the problem, either with the reduction procedure[14–19] or with the calculation of the one-hole dispersion using phenomenological parameters from the comparison with the ARPES data for $Sr_2CuO_2Cl_2$.[7–9] Here we concentrate on the connection between these two parts, providing a microscopical derivation of the extended t-J model and its direct comparison with experiment.

Following the original paper by Zhang and Rice,[14] a systematic reduction scheme from the underlying p-d model to the effective Hamiltonian describing the low-energy electronic properties of the CuO_2 plane was proposed independently by three groups of authors[15–17] and developed further in Refs. 18 and 19. The advantage of this derivation based on the cell-perturbation method is that it allows one to take into account exactly all the intracell electron correlations and provides a good convergence in treating the intercell interactions. In the present work we turn again to this problem (see Sec. II) and present in a short manner a derivation of the t-t'-t''-J Hamiltonian from the three-band p-d model containing the complete set of parameters as given by Hybertsen, Stechel, Schlüter, and Jennison.[13] Here we pursue several aims. The first one is to give the reader a compressed set of formulas connecting the parameters of the effective t-t'-t''-J model with that of the underlying p-d model. For a particular choice of the latter parameters the former ones can now be easily estimated by a numerical procedure. As for the parameter regime where the reduction is valid we refer the reader to more extensive papers.[18,19] Second, a careful analysis shows that the two versions of the reduction scheme, while being very similar to each other as they were presented originally, for instance, in Ref. 15 or in Refs. 16 and 18 contain a subtle difference. The essence of this difference remains to be discussed below. Here we notice only that within the second approach developed in Refs. 16 and 18 one encounters an ambiguity in obtaining the correct sign for the transfer-matrix elements in the effective Hamiltonian, which may lead to an incorrect band dispersion. (See also the discussion in Ref. 20.) Here we use the first approach,[15,21] which is free from this drawback. We have also deliberately chosen notation close to the one used in Refs. 18 and 20 which makes it easier to compare directly between the two approaches mentioned. In distinction to the former work,[13] we find from the reduction procedure that one has to include also a hopping term t_3 (or t'') to third nearest neighbors.

After calculating the parameters of the extended t-J-like Hamiltonian, the dispersion relation of one hole will be determined here by two different methods. The first one is based on a variational ansatz for the wave function of one hole (Sec. III) and the second one is the self-consistent Born approximation[22] (SCBA) (Sec. IV). Both of them have several advantages such that they complement each other. Here we adapt the variational approach in such a way that it allows us to investigate the influence of a three-site hopping term. This term appears naturally in the effective Hamiltonian as the second-order correction in the reduction procedure and describes also, beyond the mean-field approximation, a hole propagation accompanied by a spin-flip fluctuation. This kind of a hole motion would manifest itself as a higher-order magnon process not included usually in the conventional SCBA. On the other hand, the variational approach can hardly be applied at lower values of J and ignores also the quasiparticle lifetime effects, both of which can be done, however, within the SCBA.

The variational ansatz is proposed[23] and widely used[20,24,25] to describe the propagation of a hole as a spin polaron of minimal size, a limit that makes the problem tractable analytically. That ansatz is constructed here assuming the singlet ground state of the spin part of the Hamiltonian (Heisenberg model). We compare in detail the spin polaron dispersions corresponding to the first and second order of the reduction procedure, showing reasonable convergence. Another important question concerns the bandwidth of the quasiparticle dispersion. Recently it was claimed, using also a variational wave function, that the inclusion of additional hopping terms with the realistic range of the parameters leads to an increase of the bandwidth up to a factor of 4.[20] A result like that would contradict the experimental finding.[1] In contrast, we will show in Sec. III that a correct application of the variational ansatz leads only to a moderate increase of the calculated bandwidth in comparison to the pure t-J model.

Being restricted to a minimal size of the spin polaron, the variational ansatz is not applicable for small J. For this more realistic case we therefore use a complementary method, the SCBA. In distinction to Refs. 7 and 8 we take values for t_2 and t_3 that are calculated from ''first principles.'' Both methods (the SCBA and variational ansatz) show reasonable agreement for the calculated hole dispersion despite the fact that the SCBA uses a Néel-type two-sublattice ground state, whereas the variational ansatz does not break the spin rotational symmetry. Like in the variational approach, the bandwith calculated in the SCBA is not substantially increased due to the additional hopping integrals. Instead, a feature not visible in the pure t-J model occurs. Actually, we investigate the question of the quasiparticle damping due to the presence of additional hopping integrals (i.e., due to the bare hole dispersion) and show that this damping possesses a threshold character. Namely, if the bandwidth of the bare dispersion exceeds a certain threshold value the upper parts of the quasiparticle hole spectrum become overdamped, which results in the disappearance of quasiparticle states in some part of the Brillouin zone.

II. PROPER REDUCTION SCHEME AND THE EFFECTIVE HAMILTONIAN

We start with the three-band p-d model for the copper oxygen plane with the same parameters as those estimated by Hybertsen *et al.*[13] for the La_2CuO_4 compound. To be specific, we use the Cu-O and O-O hopping integrals $t^{pd}=1.3$ eV and $t^{pp}=0.65$ eV, respectively, the charge-transfer energy $\Delta=\varepsilon_p-\varepsilon_d=3.6$ eV, and the Coulomb repulsions given by $U_d=10.5$ eV, $U_p=4$ eV, and $U_{pd}=1.2$ eV. An effect of

TABLE I. Coefficients of the oxygen Wannier orbitals.

$\vec{i}-\vec{j}$	λ	μ	ν
0	0.958	1.454	0.0
$\vec{x}$	−0.14	−0.546	−0.266
$\vec{x}+\vec{y}$	−0.02	0.244	0.0
$2\vec{x}$	−0.014	−0.128	0.082

the direct Cu-O exchange that is not included into the original p-d model is discussed at the end of this section. To proceed with the cell-perturbation method, one has to introduce the symmetric ($p_{i\sigma}$) and antisymmetric ($\tilde{p}_{i\sigma}$) orthonormalized oxygen Wannier orbitals. The details are contained in Appendix A and in Refs. 15, 16, 18, and 19. The original Hamiltonian can now be written as

$$H=H_0+H_t+\Delta H,$$

$$H_0=\varepsilon_d\sum_{i,\sigma} n^d_{i\sigma}+\varepsilon_p\sum_{i,\sigma}(p^\dagger_{i\sigma}p_{i\sigma}+\tilde{p}^\dagger_{i\sigma}\tilde{p}_{i\sigma})+U_d\sum_i n^d_{i\uparrow}n^d_{i\downarrow},$$

$$H_t=2t^{pd}\sum_{i,j,\sigma}\lambda_{ij}(d^\dagger_{i\sigma}p_{j\sigma}+\text{H.c.})-t^{pp}\sum_{i,j,\sigma}[\mu_{ij}(p^\dagger_{i\sigma}p_{j\sigma}-\tilde{p}^\dagger_{i\sigma}\tilde{p}_{j\sigma})-\nu_{ij}(p^\dagger_{i\sigma}\tilde{p}_{j\sigma}+\text{H.c.})], \tag{2.1}$$

$$\Delta H=U_{pd}\sum_{l,i,j,\sigma}\phi_{lij}n^d_l p^\dagger_{i\sigma}p_{j\sigma}+U_p\sum_{k,l,i,j}\xi_{klij}p^\dagger_{k\uparrow}p_{l\uparrow}p^\dagger_{i\downarrow}p_{j\downarrow}+\Delta H'.$$

From now on any site index i on a square lattice labels a cell. In Eq. (2.1) the coefficients λ_{ij}, μ_{ij}, ν_{ij}, ϕ_{lij}, and ξ_{klij} follow from the Wannier transformation and fall off rapidly with the relative distance between cells. The largest values for some of these coefficients are presented in Table I. This gives us the opportunity to develop a perturbative approach: The local intracell interactions given by all relevant terms in Eq. (2.1) with coinciding lattice indices, which we denote hereafter as H_{loc}, are taken into account exactly, while the remaining intercell interactions H_{hop} are treated perturbatively.

In Eq. (2.1) the term $\Delta H'$ has minor importance for the low-energy physics. Actually, this contribution is generated by the Coulomb repulsions U_p and U_{pd} as well, but formed by antisymmetric operators $\tilde{p}_{i\sigma}$. Hence only some high-energy hole states are entangled in it. In the following we take into account exactly the local contributions contained in the two explicitly written Coulomb terms in ΔH, while the corrections due to the intercell static interactions, as well as due to $\Delta H'$, will be involved in a mean-field manner at the final stage of our derivation. It is shown below that these corrections are important to reproduce a reliable value for the superexchange constant J, but less significant for the effective hopping parameters t' and t''. Some residual Coulomb effects, not accounted here, lead to a further generalization of the t-J model involving residual interactions between doped holes (forming Zhang-Rice singlets) on neighboring cells. These interactions discussed in details in Ref. 19 are irrelevant for the present purpose of investigating a single Zhang-Rice singlet dispersion. Moreover, the effective hopping contributions coming from U_{pd} and U_p are much smaller than those from t^{pd} and t^{pp} terms. So, concerning the coefficients ϕ_{lij} and ξ_{klij}, we need only $\phi_{lll}=\phi_0$ and $\xi_{llll}=\xi_0$, which can be found to be $\phi_0=0.918$ and $\xi_0=0.211$. The sum rules $\Sigma_{l\,(\neq i)}\phi_{lii}=2-\phi_0$ and $\Sigma_{l\,(\neq i)}\xi_{llii}=\frac{1}{2}-\xi_0$ will also be used.

Further details of the cell-perturbation procedure are contained in Appendix A. In essence, after performing a diagonalization of the local Hamiltonian H_{loc}, one finds the lowest one-hole state $|if\sigma\rangle$ at a cell i and with a spin projection $\sigma=\downarrow,\uparrow$ that possesses mainly a copper character. In the insulating, undoped compound each cell is occupied by one of these spin states that interact antiferromagnetically. Among the two-hole states that appear due to doping the lowest one is the Zhang-Rice singlet denoted in the following as $|ic_1\rangle$. Excluding all high-energy hole states by the Schrieffer-Wolff transformation up to second order we obtain the effective Hamiltonian

$$H_{\text{eff}}=H_t^{(1)}+H_t^{(2)}+H_J,$$

$$H_t^{(1)}=\sum_{i\neq j,\sigma}t_{ij}X_i^{c1,f\sigma}X_j^{f\sigma,c1},$$

$$H_t^{(2)}=\sum_{j,i,j'}\left\{\sum_\sigma t^N_{jij'}X_j^{c1,f\sigma}X_{j'}^{f\sigma,c1}\hat{N}_i+\sum_{\alpha,\beta}t^S_{jij'}X_j^{c1,f\alpha}X_{j'}^{f\beta,c1}(\vec{\sigma}_{\beta\alpha}\cdot\vec{S}_i)\right\}, \tag{2.2}$$

$$H_J=\sum_{\langle ij\rangle}(J_{ij}\vec{S}_i\vec{S}_j+Y_{ij}\hat{N}_i\hat{N}_j).$$

Here $X_i^{c1,f\sigma}=|ic_1\rangle\langle if\sigma|$ is a projection operator and the density and spin operators are given as

$$\hat{N}_i=\sum_\sigma X_i^{f\sigma,f\sigma},\quad X_i^{f\sigma,f\sigma'}=|if\sigma\rangle\langle if\sigma'|,$$

$$S_i^z=\frac{1}{2}\sum_\sigma\sigma X_i^{f\sigma,f\sigma},\quad S_i^\sigma=X_i^{f\sigma,f\bar{\sigma}}, \tag{2.3}$$

with $\sigma=+\ (\uparrow)$, $-\ (\downarrow)$ and $\bar{\sigma}=-\sigma$. In Eq. (2.2) $\vec{\sigma}_{\beta\alpha}$ are matrix elements of Pauli matrices.

The results presented in Appendix A specify the parameters of the effective Hamiltonian. Namely, the first-order transfer matrix elements t_{ij} in $H_t^{(1)}$ are given by Eq. (A10) and in the framework of the t-t'-t''-J model we have to retain only the largest ones t_1, t_2, and t_3 for the lowest singlet hopping between the first, second, and third neighbors, respectively. The resulting three-site transfer-matrix elements including a spin-dependent hopping $t^{N/S}_{jij'}$ in $H_t^{(2)}$ are given by Eq. (A14). Again we keep only the largest contributions with the obvious notation $t_2^{N/S}=t^{N/S}_{j,j+x,j+x+y}$, $t_3^{N/S}=t_{j,j+x,j+2x}$. Finally, the parameters of the exchange part H_J are presented by Eq. (A16). Using the parameters of the p-d model in accordance with Hybertsen *et al.*,[13] we obtain the values of the above parameters as given in Table II. Because the exchange between more distant cells is small, we restrict ourselves to the nearest neighbors $J_1=J$.

Though the form of the Hamiltonian (2.2) coincides with the one derived in Refs. 18 and 20 the values of its parameters calculated with the same parameters of the underlying

TABLE II. Hopping and exchange parameters in first and second order of the reduction procedure.

Parameter	$i=1$	$i=2$	$i=3$
t_i (meV)	498	-41	77
J_i (meV)	235	-0.2	4
t_i^N (meV)		25	12
t_i^S (meV)		-58	-58

p-d model may be different. The difference reduces mainly to a sign problem. Actually, as an intermediate step of the present derivation we define the action of the original operators $A_{i\sigma}=d_{i\sigma}$, $p_{i\sigma}$, and $\tilde{p}_{i\sigma}$ within the ''new'' one-cell basis $|a\rangle$ (see Appendix A) as

$$A_{i\sigma}=\sum_{a',a}\langle a'|A_\sigma|a\rangle X_i^{a'a} \tag{2.4}$$

and then substitute these expansions into the intercell part of the Hamiltonian $H_{\rm hop}$. Because the coefficients in Eq. (2.4) are unambiguously connected with the matrix elements of unitary transformations (A8) as $\langle a'|A_\sigma|a\rangle = \Sigma_{\nu'\nu}\langle a'|\nu'\rangle\langle\nu'|A_\sigma|\nu\rangle\langle\nu|a\rangle$ and the decomposition of A_σ given by $\langle\nu'|A_\sigma|\nu\rangle$ within the ''old'' basis $|\nu\rangle$ is unique, the two-site matrix elements $t_{ij,\sigma}$ and $\tilde{t}_{ij,\sigma}$ in Eq. (A9) are also obtained in a unique fashion. In another approach[16,18] one first defines a set of two-cell wave functions as direct products such as $|ia,jb\rangle=|ia\rangle\otimes|jb\rangle$. As it was pointed out in Ref. 20 an evaluation of the matrix elements $\langle ia',jb'|H_{\rm hop}|ia,jb\rangle$ within this basis contains an uncertainty in the signs for these elements, which requires a special subtle convention to fix the signs.

Further, to define properly the exchange constant J, each pair of sites $\langle ij\rangle$ enters into the summation in H_J [Eq. (2.2)] only once, which explains why the calculated value of $J=235$ meV is twice as large as the one presented in Ref. 18. Our estimation so far is nearly the same as the one reported in Ref. 19 and significantly larger than the accepted experimental value for copper oxides. Several corrections of the opposite (ferromagnetic) sign to J, however, should be taken into account. Two sorts of corrections are mainly due to intercell Coulomb interactions presented by the off-site part of ΔH in Eq. (2.1) where one has to distinguish between the density-density and the spin-dependent exchange terms.[19] The former one, as well as the residue $\Delta H'$, we treat in a mean-field manner as presented at the end of Appendix A. The procedure that is explained there leads to a substantial reduction of the nearest-neighbor exchange constant J to a value of 202 meV. The other parameters of Table II are only slightly changed (t_2 to a value of -36 meV, for instance), which we therefore neglect in the following. Similar effects of suppression of the superexchange due to the intersite Coulomb interaction in the charge-transfer system was recently discussed by Eder *et al.*[26] The Coulomb exchange between neighboring cells induced by U_p and considered in Refs. 18 and 19 leads to the second correction

$$\Delta J_p=-2\xi_1 v^4 U_p. \tag{2.5}$$

The third correction stems from the direct Cu-O exchange

$$H_K=-2K\sum_{\langle il\rangle}\vec{S}_i^{(d)}\cdot\vec{s}_l^{(p)}, \tag{2.6}$$

where $K=0.18$ eV (Refs. 13 and 27) and the spin operators $\vec{S}_i^{(d)}$ and $\vec{s}_l^{(p)}$ are defined in terms of the original $d_{i\sigma}$ and $p_{l\sigma}$ ($\vec{l}=\vec{\imath}\pm\vec{x}/2$, $\vec{\imath}\pm\vec{y}/2$) operators. This interaction, not included in the original p-d model, is considered by several authors.[13,27,28] Within the reduction procedure developed here the correction to the exchange constant is

$$\Delta J_K=-4\phi_1 u^2 v^2 K. \tag{2.7}$$

In Eqs. (2.5) and (2.7) $\xi_1\simeq 0.06$ and $\phi_1=0.243$ are coefficients of the Wannier transformation and u and v are the components of the T_1 matrix (A8). So for the present parameters, the calculation gives $\Delta J_p=-33$ meV and $\Delta J_K=-34$ meV. Thus we can confirm the observation of Stechel and Jennison[13,27] that the inclusion of the direct Cu-O exchange (2.6) leads to a considerable reduction of the value of J. Taking into account that the exchange (2.6) diminishes the energy distance between the bare singlet and triplet states [see Eq. (A6)] by an amount of 2 K leads to an additional correction of -5 meV. The resulting superexchange constant then becomes $J=130$ meV, which is close to the one usually reported for copper oxides and coincides with the estimation obtained by Hybertsen *et al.*[13]

III. VARIATIONAL ANSATZ FOR THE ONE-HOLE MOTION

Now we use the Hamiltonian (2.2) with the parameters given in Table II to calculate the dispersion of one hole in the antiferromagnetic state of the Heisenberg Hamiltonian H_J (with only nearest-neighbor exchange). For that purpose we may set the density operator $\hat{N}_i$ equal to unity. From now on we neglect also the constant energy shift resulting from the $\hat{N}_i$ in H_J [Eq. (2.2)]. For convenience we write the three-site term $H_t^{(2)}$ in a more spin rotational invariant form and find

$$H_t^{(1)}+H_t^{(2)}=\hat{K}+\hat{T}\quad,\quad \hat{K}=\sum_{i,b,\sigma}\tilde{t}_b X_i^{0\sigma}X_{i+b}^{\sigma 0}, \tag{3.1}$$

with

$$\tilde{t}_b=\begin{cases}\tilde{t}_1=t_1 & \text{if } b \text{ is a nearest-neighbor vector}\\ \tilde{t}_2=t_2+2(t_2^N-t^S/2) & \text{if } b \text{ is a second-nearest-neighbor vector}\\ \tilde{t}_3=t_3+(t_3^N-t^S/2) & \text{if } b \text{ is a third-nearest-neighbor vector}\\ 0 & \text{otherwise,}\end{cases} \tag{3.2}$$

where we replace $X_i^{c1,f\sigma}=X_i^{0\sigma}$ to simplify the notation. So the $\hat{K}$ term involves, on the mean-field level, also the three-site hopping term. With the parameters presented in Table II the effective hopping integrals $\tilde{t}_2$ and $\tilde{t}_3$ take the values

$$\tilde{t}_2=66 \text{ meV}, \quad \tilde{t}_3=118 \text{ meV}. \tag{3.3}$$

The remaining part $\hat{T}$ of the three-site term has the form

$$\hat{T}=t^S \sum_{i,a_1,a_2,\sigma_1,\sigma_2} \overline{\delta_{a_1,-a_2}} X_i^{0\sigma_1} X_{i+a_1}^{\sigma_1\sigma_2} X_{i+a_1+a_2}^{\sigma_2 0}, \tag{3.4}$$

with $t^S=t_2^S=t_3^S$ as given in Table II, where the sum over a_1,a_2 goes over next nearest neighbors and $\overline{\delta_{a_1,-a_2}}$ is zero if $a_1=-a_2$ and equal to unity in all the other cases. The $\hat{T}$ term involves contributions to a hole propagation that are beyond the mean-field approximation. Actually, in the slave-fermion linear spin-wave representation developed in Sec. IV, this term corresponds to two-magnon scattering processes that couple to the moving hole. It requires some extension of the conventional SCBA to take this kind of process into account.[8]

Below we adapt the variational ansatz[24,25] in a form that allows us to calculate the influence of the term $\hat{T}$ in detail. The variational ansatz is constructed with the spin singlet ground state $|\Psi_0)$ of the Heisenberg Hamiltonian. The first basis function corresponds to the creation of one hole in $|\Psi_0)$, i.e., $\phi_i^{0^+}|\Psi_0)=X_i^{0\sigma}|\Psi_0)$, and the other four basis operators

$$\phi_i^a=\sum_s X_{i-a}^{\sigma s} X_i^{s0}, \quad a=1,\ldots,4 \tag{3.5}$$

involve a spin defect at the neighboring site a of the hole and correspond to the so-called string state of length one. Here and in the following a denotes at the same time a neighboring vector and a number between 1 and 4 that are connected with each other in a unique way. Thus the ansatz corresponds to a magnetic polaron of minimal length. It describes states with a total spin $S_{\text{tot}}=1/2$ that are known to represent the lowest states of the t-J model with one hole.[3,4]

To calculate analytically the dispersion of the lowest one-hole state, we use the ansatz (3.5) like in the Ritz variational principle. This means that we determine the overlap matrix

$$S^{ab}=\sum_j (\Psi_0|\phi_l^a \phi_j^{b^+}|\Psi_0) e^{ik(j-l)} \tag{3.6}$$

and the Hamilton matrix

$$H^{ab}(k)=\sum_j (\Psi_0|[\phi_l^a,\hat{H}]\phi_j^{b^+}|\Psi_0) e^{ik(j-l)}, \tag{3.7}$$

where a and b denote different basis functions $(a,b=0,\ldots,4)$ and the Hamiltonian (3.7) contains the kinetic and exchange energies as well as the three-site terms $\hat{H}=\hat{K}+\hat{T}+\hat{H}_J$.

The matrix elements of $\hat{K}$ and $\hat{H}_J$ have already been given in Ref. 25, where the influence of doping on the spectrum had been studied. The doping was simulated by temperature or a frustration term in the spin part of the Hamitonian.

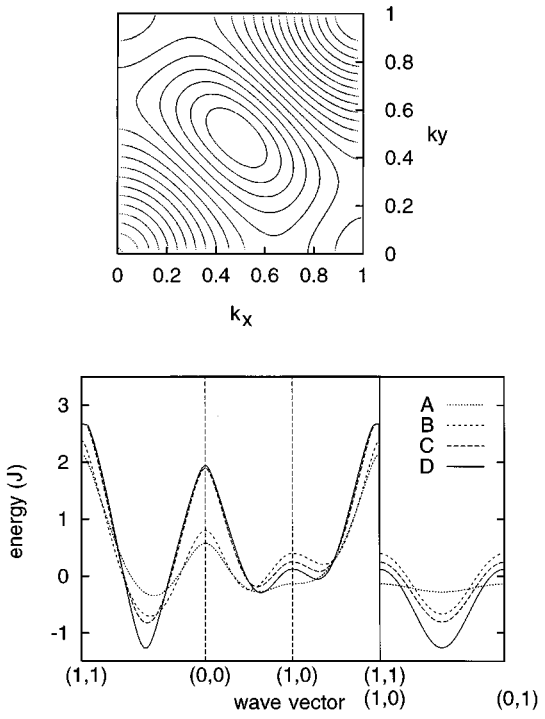

FIG. 1. Quasiparticle dispersion within the variational ansatz for the four sets of parameters A–D, which are explained in the text, and a contour plot for case D. The wave vectors are given in units of (π,π) (lattice constant $a=1$).

Therefore, we will not repeat these matrices here. The only new contribution, the matrix of the $\hat{T}$ term, is given in Appendix B. All the contributions to the Hamilton matrix $\underline{H}$ [Eq. (3.7)], as well as the overlap matrix $\underline{S}$, depend on the ground state $|\Psi_0)$ of the pure spin system due to static spin-spin correlation functions. Here we approximate four- and six-point spin-spin correlation functions by decoupling them into two-point spin-spin correlation functions.[29] These have been calculated by a self-consistent, spin rotational invariant Green's-function technique for the Heisenberg model.[30] We have checked that the modified spin-wave theory of Takahashi[31] gives essentially the same results. Once we have determined the spin-spin correlation functions, we can calculate the quasiparticle dispersion by finding the lowest eigenvalue of $\underline{HS}^{-1}$.

In Fig. 1 we compare the results for four sets of parameters. Set A corresponds to the pure t-J model; in set B we take the additional hopping terms t_2 and t_3, calculated in first order (Table II); in set C we incorporate also the second-order corrections in the mean-field approximation (3.3), and in the complete case, set D, the term $\hat{T}$ describing the three-site hopping beyond the mean-field level is added. In all cases we choose $J=t_1$ $(=t)$ since our variational ansatz is restricted to a magnetic polaron of minimal radius and is not applicable at smaller J. For a proper treatment of the more realistic case of smaller values of J one needs to extend the basis (3.5). Nevertheless, it is worth discussing how the results change if one retains the restricted basis even for $J<t$. In fact, we found that the shape of the dispersion does not change and the bandwidth stays nearly the same as in Fig. 1. That is in contrast to the general expectation that a

decrease of J/t leads to a decrease of the bandwidth. Such a decrease is not reproduced in the present variational ansatz. The scaling of the bandwidth with the value of J is already known for the pure t-J model for a long time[2–6] and the results of Sec. IV prove it also for the case with additional hopping terms on one sublattice (see also Refs. 25 and 32). Therefore, we feel justified to present the results in Fig. 1 in units of E/J, suggesting a simple scaling to smaller values of J.

In case A without additional hopping terms the energies at $(\pi/2,\pi/2)$ and $(\pi,0)$ are nearly degenerate; there is very little dispersion along $(\pi,0)$-$(0,\pi)$ and, consequently, a highly anisotropic minimum at $(\pi/2,\pi/2)$. These results are consistent with former calculations for the pure t-J model.[2–6] In Refs. 25 and 32 we found that the variational ansatz in the form presented here agrees well with the exact diagonalization data of a 4×4 cluster with periodic boundary conditions. The difference in energy between (π,π) and $(0,0)$ is due to our spin rotational invariant procedure, which does not break the lattice translational symmetry. The bandwidth is $2.5J$ and is slightly enhanced in comparison to the SCBA and the exact diagonalization,[3,4] which may be due to our limited basis. One should note that the discrepancies between different methods is especially large at (π,π), but smaller at $(0,0)$ and at the band bottom.

In all the other cases $(B$–$D)$ taking into account the details of the reduction procedure with increasing sophistication we observe the common effect that the minimum at $(\pi/2,\pi/2)$ becomes a more isotropic one. That is in agreement with the general tendency that is experimentally[1,10] observed in $Sr_2CuO_2Cl_2$. In Fig. 1 one notes a small anisotropy (the ratio between effective masses in the extremal directions) between 1.2 (case B) and 2 (case D). This feature is closer to the results in Ref. 10 than to that in Ref. 1, where a highly isotropic minimum was reported. Although the approximate degeneracy for the hole energies at $(\pi/2,\pi/2)$ and $(\pi,0)$ is now lifted out, our results fail to reproduce the dispersionless feature along the line $(0,0)$-$(\pi,0)$ seen in one of the experiments.[1]

To interpret our result let us note that the additional hopping terms t_2 ($\tilde{t}_2$) and t_3 ($\tilde{t}_3$) act in such a way that they suppress the motion in the direction of the copper-oxygen bond, but facilitate the motion along the diagonal. We can support the observation of Ref. 33, pointing to the importance of a particular combination of the additional hopping parameters, namely, t_2-2t_3 (or, correspondingly, $\tilde{t}_2-2\tilde{t}_3$ in cases C and D). This combination is of crucial importance for the shape of the hole band and is responsible, for instance, for the dispersion along the boundary of the antiferromagnetic (AFM) Brillouin zone (BZ). Comparing sets B and C (or D), one can see that the value of the above combination does not change significantly, though the t_2 parameter changes sign: $t_2<0$, while $\tilde{t}_2>0$. This explains the weak difference between the dispersions calculated for the B and C cases. Comparing cases D and C, we observe that the detailed consideration of the nontrivial part $\hat{T}$ of the three-site term has little influence on the calculated dispersion relation. In case D the bandwith is enhanced due to the additional hopping terms by a factor of 1.5 as compared to the pure t-J model. That is much less than what is reported in Ref. 20. The reason may be the difference in the variational ansatz.

IV. SELF-CONSISTENT BORN APPROXIMATION

A powerful method of investigating a hole dispersion in an AFM background is the SCBA applied to the t-J-like model in slave-fermion representation

$$X_i^{\sigma 0}=f_i b_{i\sigma}^{\dagger}. \tag{4.1}$$

Here f_i is the spinless fermion associated with the charge degrees of freedom and the Schwinger boson operator $b_{i\sigma}^{\dagger}$ keeps track of the spins. In the linear spin wave approximation and after the Fourier transformation the Hamiltonian (2.2) can be written as

$$H_t^{(1)}=\sum_k \varepsilon_k^{(1)} f_k^{\dagger} f_k+\sum_{k,q} M(k,q)[f_k^{\dagger} f_{k-q}\beta_q+\text{H.c.}]+\delta H_t^{(1)},$$

$$H_t^{(2)}=\sum_k \varepsilon_k^{(2)} f_k^{\dagger} f_k+\delta H_t^{(2)}, \tag{4.2}$$

$$H_J=\sum_q \omega_q \beta_q^{\dagger}\beta_q+\Delta E_J^0, \quad \omega_q=szJ\sqrt{1-\gamma_q^2}.$$

Here the lowest-order hole-magnon vertex $M(k,q)$ has the form

$$M(k,q)=\frac{zt_1}{\sqrt{N}}(u_q\gamma_{k-q}+v_q\gamma_k), \quad \gamma_k=\frac{1}{2}(\cos k_x+\cos k_y), \tag{4.3}$$

with u_q and v_q being the coefficients of the Bogoliubov transformation for spin variables. ΔE_J^0 is the change of the magnetic ground-state energy due to introducing a hole into the half-filled background ($s=\frac{1}{2}$, $z=4$):

$$\Delta E_J^0=Jz\left[s^2+s\left(1-\frac{1}{N}\sum_q \sqrt{1-\gamma_q^2}\right)\right]. \tag{4.4}$$

We have to define also

$$\varepsilon_k^{(1)}=4t_2\cos k_x\cos k_y+2t_3[\cos 2k_x+\cos 2k_y] \tag{4.5}$$

and $\varepsilon_k^{(2)}$ takes the same form as $\varepsilon_k^{(1)}$ with the replacement $t_2\to 2(t_2^N-\frac{1}{2}t_2^S)$, $t_3\to(t_3^N-\frac{1}{2}t_3^S)$. The main differences of the Hamiltonian (4.2) from the well-known spin polaron model arising from the pure t-J model are (i) the appearance of the bare dispersions $\varepsilon_k^{(1)}$ and $\varepsilon_k^{(2)}$ due to free hole hopping processes of the first $(\sim t_2,t_3)$ and second $(\sim t_2^{N/S},t_3^{N/S})$ order and (ii) the involvement in the hole-magnon interaction of two-magnon processes, such as $f_k^{\dagger}f_{k-q_1-q_2}\beta_{q_1}\beta_{-q_2}^{\dagger}$, which are included in the first-order $\delta H_t^{(1)}$ $(\sim t_2,t_3)$ and the second-order $\delta H_t^{(2)}$ $(\sim t_2^{N/S},t_3^{N/S})$ corrections with their own more complicated vertices not written here.

The main part of the hole-magnon interaction $(\sim t=t_1)$ is given by the second term in $H_t^{(1)}$. As it was proved in numerous investigations for the pure t-J model, just this term is responsible for the formation of a quasiparticle (QP) hole propagation with a bandwidth $W\simeq 2J$ in the wide range of

J values if $J<t$. Recently Bala, Oleś, and Zaanen[8] examined an extended t-J-like model and showed numerically that the two-magnon processes mentioned above are insignificant, while the influence of a bare hole dispersion on the overall picture resulting from the SCBA is important. Similar though more restricted results were also reported by Nazarenko *et al.* in Ref. 7 based on the t-t'-J model not including three-site hopping terms.

We addressed the same problem and calculated within the SCBA a single hole (spin polaron) band dispersion based on the Hamiltonian (4.2), however, without $\delta H_t^{(1)}$ and $\delta H_t^{(2)}$, i.e., higher-order magnon processes are assumed to be insignificant. We compare the results with that derived in the pure t-J model[6] to highlight the effects of free hole propagation given by a bare dispersion $\varepsilon(k)$. We differentiate the case where $\varepsilon(k)=\varepsilon_k^{(1)}$ and a more complete case where the second-order hopping processes also contribute, $\varepsilon(k)=\varepsilon_k^{(1)}+\varepsilon_k^{(2)}$.

We solved numerically on the 32×32 cluster the equation for the self-energy

$$\Sigma(\vec{k},\omega)=\sum_q M^2(\vec{k},\vec{q})G(\vec{k}-\vec{q},\omega-\omega_q)$$

with the hole Green's function

$$G(\vec{k},\omega)=\frac{1}{\omega-\varepsilon(\vec{k})-\Sigma(\vec{k},\omega)}.$$

The organization of the self-consistent numerical procedure is the same as in Ref. 34 and the exchange integral is chosen to be either $J=0.4t$ or $J=t$.

Let us now discuss the results of numerical calculations. The upper panels in Figs. 2(a) and 2(b) show the spectral density function $A(k,\omega)=-\pi^{-1}\mathrm{Im}G(k,\omega+i\eta)$ for two momenta $\vec{k}=(\pi/2,\pi/2)$ and $\vec{k}=(0,0)$ as the most representative ones. Here we set $J=0.4t$ and $\varepsilon(k)=\varepsilon_k^{(1)}+\varepsilon_k^{(2)}$, which corresponds to a bare bandwidth $W'\simeq 3J$. One can see at $\vec{k}=(\pi/2,\pi/2)$ a very sharp, well-pronounced peak in the low-energy part of the spectrum, while at $\vec{k}=(0,0)$ this peak is strongly broadened. To get more insight into the problem, we show also, in the lower panels, the imaginary part of the self-energy $\mathrm{Im}\Sigma(k,\omega)$. [Because $A(\vec{k},\omega)\geqslant 0$ and $\mathrm{Im}\Sigma(k,\omega)\leqslant 0$ these two pieces can be easily distinguished in the pictures.] It can be seen from Fig. 2(a) (and we examined it numerically) that at $\vec{k}=(\pi/2,\pi/2)$ in the energy interval where the low-energy peak is located $\mathrm{Im}\Sigma(k,\omega)\simeq 0$ holds with good accuracy. So the visible broadening of this well-defined QP peak is due to the artificial value $\eta=0.005t$ introduced in our numerical procedure to facilitate the computations. At the same time, at $\vec{k}=(0,0)$ the position of the low-energy peak coincides with a large value of $\mathrm{Im}\Sigma(k,\omega)$ and hence this peak can hardly be detached from the incoherent part of the spectrum. In this way we verified the existence of a QP hole state in the most part of the AFM BZ except for the center $\vec{k}=(0,0)$ and its nearest vicinity, where the QP is not well defined. That is the effect of a bare dispersion $\varepsilon(k)$ with a large enough bandwidth W'. We notice that in the pure t-J model $\varepsilon(k)=0$, a well-defined QP state, though with a weight at $\vec{k}=(0,0)$ somewhat smaller than at $\vec{k}=(\pi/2,\pi/2)$, exists in the whole BZ. Bala *et al.*[8] came to the same conclusion. Further support of this statement was found by taking the value $J=t$. In this case for any dispersion $\varepsilon(k)$ examined here, the bare bandwidth is not too high, $W'<1.2J$, and the well-defined QP state is found in the whole BZ.

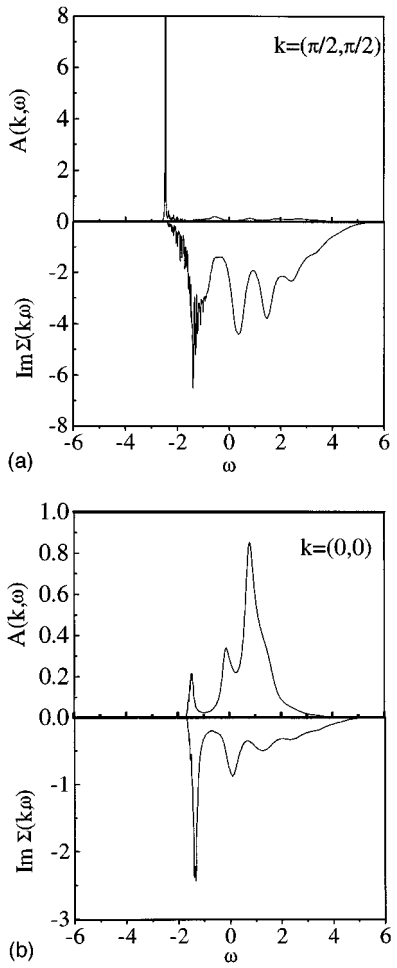

FIG. 2. Spectral density and imaginary part of the self-energy for $J=0.4t$, $t'/t=0.13$, and $t''/t=0.24$ ($t=1$) within the SCBA at (a) $(\pi/2,\pi/2)$ and (b) (0,0).

The resulting dispersion $E(k)$ extracted from the low-energy peak positions in $A(\vec{k},\omega)$ is shown in Figs. 3(a) and 3(b) for $J=t$ and $J=0.4t$, respectively. Three curves are plotted in each picture to compare the following possibilities: (a) the pure t-J model $\varepsilon(k)=0$, (b) the t-t'-t''-J model with a reduced bare dispersion $\varepsilon(k)=\varepsilon_k^{(1)}$, and (c) the full version for the bare dispersion $\varepsilon(k)=\varepsilon_k^{(1)}+\varepsilon_k^{(2)}$. In Fig. 3 the continuous thin line is reserved for case (a), while the circles and crosses are for (b) and (c), respectively. Please note also the agreement of the absolute values of the energies between the variational ansatz (Fig. 1) and the SCBA for $J=t$ [Fig. 3(a)]. For that it is important to include the energy shift ΔE_J^0 [Eq. (4.4)] (which was done in Fig. 3, but not in Fig. 2). Only those circles (crosses) that are connected by a thick (thin) line in Fig. 3 correspond to well-defined QP states. For $J=0.4t$ [Fig. 3(b)], well-defined QP states do not occur on the upper parts of the dispersions, i.e., at the center

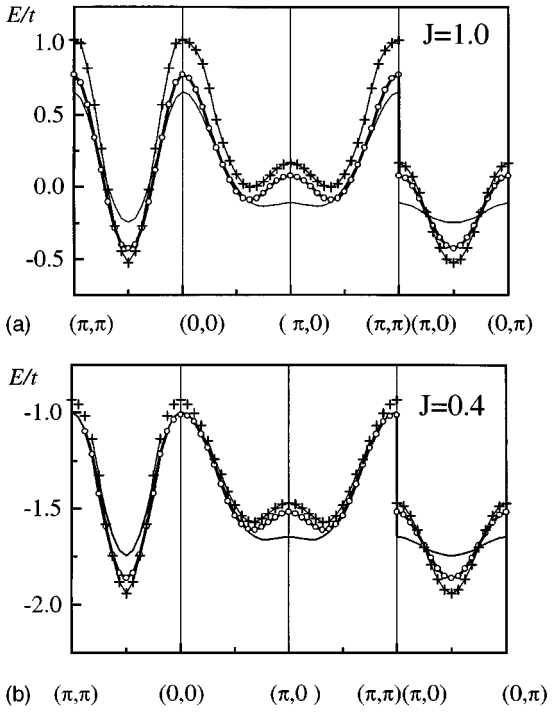

FIG. 3. Quasiparticle dispersion within the SCBA for the parameter sets A (thin line), B (circles and thick line), and C (crosses and thin line) for (a) $J=t$ and (b) $J=0.4t$. Only those points that are connected by a line have a well-defined QP character.

$\vec{k}=(0,0)$ and at the equivalent points $\vec{k}=(\pm\pi,\pm\pi)$ of the BZ, where the QP states are overdamped for both cases (b) and (c). So the resulting true QP bandwidth is estimated to become $W\simeq 2J$, not larger. Actually, high-energy QP states lost their identity due to a strong scattering into hole states of lower energy with emitting a magnon. This scattering process possesses a threshold character and becomes much more intensive if the bandwidth of QP states tends to exceed the maximum magnon energy $2J$. Some proof of this statement is given in Appendix C.

Let us note that the effective hopping parameters t' and t'' can be defined as $t'=t_2+2(t_2^N-\frac{1}{2}t_2^S)$ and $t''=t_3+(t_3^N-\frac{1}{2}t_3^S)$. Just these parameters determine a bare hole dispersion $\epsilon(k)=\epsilon_k^{(1)}+\epsilon_k^{(2)}$ with the values estimated to be $t'/t=0.13$ and $t''/t=0.24$ (Table II). Recently, Belinicher, Chernyshev, and Shubin reported[35] the results of the SCBA calculation for the QP dispersion based on the same Hamiltonian (1.2) with $J/t=0.4$, but for another choice of the t' and t'' parameters: $t'/t\approx 0$ and $t''/t=0.25$; the latter is very close to our estimate. The authors in Ref. 35 found nearly flat QP dispersion along the line $(0,0)$-$(\pi/2,0)$-$(\pi,0)$ and almost isotropic dispersion near the QP band minimum. So it was claimed in Ref. 35 that excellent agreement between the SCBA computation and ARPES results of Ref. 1 is reached.

Our SCBA computation performed with the same values of parameters t' and t'' as in Ref. 35 and $J/t=0.4$ show, however, that the relevant QP dispersion curve depicted in Fig. 3(b), case (c), only slightly changes and preserves its main character. Namely, the QP energies at $(\pi,0)$ remain nearly in the middle of the band and the effective QP mass in the $(\pi,0)$-$(0,\pi)$ direction is about twice as large as that in the $(0,0)$-(π,π) direction. So we may infer that, to a large extent, the overall shape of the QP dispersion is insensitive to the t' value provided that there is a large t'' parameter.

V. SUMMARY AND CONCLUSIONS

To investigate the hole band dispersion in copper oxides we have derived the form and estimated the parameter values for a one-band extended t-J model as the low-energy limit of the underlying three-band p-d model for the CuO_2 plane.[13] The reduction procedure is based on the cell-perturbation method and developed here in such a way that there is no ambiguity in the signs of the hopping integrals in the effective one-band model. In distinction to former expectations,[7,13] the reduction procedure pointed to the necessity to include not only a hopping to second (t'), but also to third (t'') neighbors.

Further, in calculating the hole band dispersion, both within the variational approach and the SCBA, we distinquished between two sets of parameters derived in first ($t'/t=-0.08$, $t''/t=0.15$) and up to second order ($t'/t=0.13$, $t''/t=0.24$). The leading effect of the so-called three-site hopping term was shown to be provided already on the mean-field level. Both methods and both sets of parameters were shown to lead to a similar shape of the hole dispersion.

As the main result we found that the present extension of the t-J model leads to a more isotropic minimum at $(\pi/2,\pi/2)$ in comparison to the pure t-J model. The calculated dispersion between $(0,0)$ and $(\pi,0)$ does not completely vanish (in distinction to the dispersionless feature in Ref. 1) and it remains an anisotropy at $(\pi/2,\pi/2)$ between 1.2 and 2. It is interesting to note that also the more recent experiment[10] indicated a small anisotropy around the minimum $(\pi/2,\pi/2)$.

Let us briefly discuss the limitations imposed by our approximations or in the present t-t'-t''-J model itself. For instance, the higher-order magnon processes coupled to the hole propagation and generated by the additional hopping terms deserve to be examined more carefully in a refined SCBA. The form of the present effective model seems to be rather general; however, the values of its parameters, as well as that for the p-d model, may vary for different copper oxides. In the present paper we used parameters that were derived for La_2CuO_4.[13] So a constrained density-functional calculation for $Sr_2CuO_2Cl_2$ is strongly desirable. In that case it might be helpful that we gave the formulas of the reduction procedure explicitly.

The experiments[1,10] indicated also that the quasiparticle states on the upper part of the hole spectrum are not so well defined as at the bottom. Along with this finding, our results (see also Ref. 8) show a threshold character of this quasiparticle damping. Actually, an increase of the resulting hole dispersion, due to the presence of a bare hole band of considerable bandwidth, is inhibited by strong scattering of the high-lying quasiparticle states into lower ones. That imposes an upper cutoff, roughly $2J$, on the resulting hole bandwidth and leads to the disappearence of well-defined quasiparticle states in considerable regions of the Brillouin zone centered at $(0,0)$ and (π,π). On the other hand, our calculations re-

sult in a well-defined quasiparticle state around $(\pi,0)$, where it is strongly overdamped in the experiment.[1,10] The above-described mechanism of damping due to a large bare dispersion would provide us with such an effect, however, only for unrealistic values of the parameters t' and t''. Therefore, it seems likely that additional damping processes, not included in the SCBA, exist as well.

ACKNOWLEDGMENTS

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APPENDIX A

In this appendix the main steps of the mapping from the p-d to the effective t-t'-t''-J model are presented. We start with the definition of the symmetric and antisymmetric ortonormalized oxygen orbitals by introducing the corresponding annihilation operators $p_{q\sigma}$ and $\tilde{p}_{q\sigma}$, respectively,[16]

$$p_{q\sigma}=\frac{is_{q,x}}{\lambda_q}p^{(x)}_{q\sigma}-\frac{is_{q,y}}{\lambda_q}p^{(y)}_{q\sigma},$$

$$\tilde{p}_{q\sigma}=\frac{is_{q,y}}{\lambda_q}p^{(x)}_{q\sigma}+\frac{is_{q,x}}{\lambda_q}p^{(y)}_{q\sigma}. \tag{A1}$$

Here $p^{(x,y)}_{q\sigma}$ are the Fourier-transformed original oxygen orbitals

$$p^{(\alpha)}_{q\sigma}=\frac{1}{\sqrt{N}}\sum_i p^{(\alpha)}_{\vec{i}+\vec{\alpha}/2,\sigma}e^{-i\vec{q}(\vec{i}+\vec{\alpha}/2)}, \tag{A2}$$

where $\vec{\alpha}=\vec{x},\vec{y}$ are two orthogonal unit vectors on the square lattice with sites $\vec{i}$ (lattice constant $a=1$).The coefficients $s_{q,\alpha}$ and λ_q are defined below.

The p-d model originally written in terms of $p^{(x)}_{l,\sigma}$ and $p^{(y)}_{l,\sigma}$ operators ($\vec{l}=\vec{i}+\vec{\alpha}/2$) can now be presented in terms of Wannier orbitals associated with $p_{i\sigma}$ and $\tilde{p}_{i\sigma}$ [Eq. (2.1)] in the main text. This can be done first by inverting the relation (A1) and then performing the Fourier transformation to the coordinate space. This transformation generates a set of coefficients λ_{ij}, μ_{ij}, and ν_{ij} defined as

$$\{\lambda,\mu,\nu\}(\vec{i}-\vec{j})=\frac{1}{N}\sum_q \{\lambda,\mu,\nu\}_{\vec{q}}e^{i\vec{q}(\vec{i}-\vec{j})},$$

$$\lambda_q^2=s_{q,x}^2+s_{q,y}^2, \quad \mu_q=8\frac{s_{q,x}^2s_{q,y}^2}{\lambda_q^2},$$

$$\nu_q=4s_{q,x}s_{q,y}(s_{q,x}^2-s_{q,y}^2)/\lambda_q^2, \tag{A3}$$

$$s_{q,x}=\sin(q_x/2), \quad s_{q,y}=\sin(q_y/2).$$

These coefficients fall off rapidly with the distance $(\vec{i}-\vec{j})$, Table I. A brief remark should be made on the symmetry of the coefficients. Actually, λ_{ij} and μ_{ij} do not change but ν_{ij} does change sign when $(\vec{i}-\vec{j})$ is reflected in the $[1,1]$ or $[1,\overline{1}]$ direction. In particular, $\nu(\vec{y})=-\nu(\vec{x})$, which leads to opposite second-order corrections for the effective hole hopping amplitudes to the second $[\sim\nu(x)\nu(y)=-\nu^2(x)]$ and third $[\sim\nu^2(x)]$ neighbors.

Besides the vacuum $|i0\rangle$ and one-hole states $|id\sigma\rangle\equiv d^\dagger_{i\sigma}|0\rangle$, $|ip\sigma\rangle\equiv p^\dagger_{i\sigma}|0\rangle$ and $|i\tilde{p}\sigma\rangle\equiv\tilde{p}^\dagger_{i\sigma}|0\rangle$, one has to introduce a two-hole basis for each cell (the site index $\vec{i}$ is implied) as

$$|\psi_1\rangle=S(d^\dagger,p^\dagger)|0\rangle, \quad |\tau m\rangle=T_m(d^\dagger,p^\dagger)|0\rangle,$$

$$|\psi_2\rangle=p^\dagger_\uparrow p^\dagger_\downarrow|0\rangle,$$

$$|\psi_3\rangle=d^\dagger_\uparrow d^\dagger_\downarrow|0\rangle, \tag{A4}$$

$$|\tilde{\psi}_1\rangle=S(d^\dagger,\tilde{p}^\dagger)|0\rangle, \quad |\tilde{\tau}_1 m\rangle=T_m(d^\dagger,\tilde{p}^\dagger)|0\rangle,$$

$$|\tilde{\psi}_2\rangle=S(p^\dagger,\tilde{p}^\dagger)|0\rangle, \quad |\tilde{\tau}_2 m\rangle=T_m(p^\dagger,\tilde{p}^\dagger)|0\rangle,$$

$$|\tilde{\psi}_3\rangle=\tilde{p}^\dagger_\uparrow\tilde{p}^\dagger_\downarrow|0\rangle,$$

where

$$S(a^\dagger,b^\dagger)=\frac{1}{\sqrt{2}}(a^\dagger_\uparrow b^\dagger_\downarrow-a^\dagger_\downarrow b^\dagger_\uparrow),$$

$$T_m(a^\dagger,b^\dagger)=\left(a^\dagger_\uparrow b^\dagger_\uparrow,\frac{1}{\sqrt{2}}(a^\dagger_\uparrow b^\dagger_\downarrow+a^\dagger_\downarrow b^\dagger_\uparrow),a^\dagger_\downarrow b^\dagger_{-\downarrow}\right) \tag{A5}$$

for $m=+1,0,-1$, respectively. The energies of these states are given by

$$\varepsilon_{\psi 1}=\varepsilon_d+\varepsilon_p-t^{pp}\mu_0+U_{pd}\phi_0, \quad \varepsilon_\tau=\varepsilon_{\psi 1}$$

$$\varepsilon_{\psi 2}=2(\varepsilon_p-t^{pp}\mu_0)+U_p\xi_0,$$

$$\varepsilon_{\psi 3}=2\varepsilon_d+U_d, \tag{A6}$$

$$\varepsilon_{\tilde{\psi}1}=\varepsilon_d+\varepsilon_p+t^{pp}\mu_0, \quad \varepsilon_{\tilde{\tau}1}=\varepsilon_{\tilde{\psi}1}$$

$$\varepsilon_{\tilde{\psi}2}=2\varepsilon_p, \quad \varepsilon_{\tilde{\tau}2}=\varepsilon_{\tilde{\psi}2}$$

$$\varepsilon_{\tilde{\psi}3}=2(\varepsilon_p+t^{pp}\mu_0).$$

The three- and four-hole states do not contribute to the low-energy physics we are interested in. Within this basis the Hamiltonian can now be written as $H=H_{\rm loc}+H_{\rm hop}$, where the zeroth-order Hamiltonian $H_{\rm loc}$ presents the intracell interaction

$$\begin{aligned}H_{\rm loc}=&\sum_{i,\sigma}\{\varepsilon_d X_i^{d\sigma,d\sigma}+(\varepsilon_p-t^{pp}\mu_0)X_i^{p\sigma,p\sigma}+t(X_i^{d\sigma,p\sigma}\\&+\text{H.c.})\}+\sum_{i,\sigma}(\varepsilon_p+t^{pp}\mu_0)X_i^{\tilde p\sigma,\tilde p\sigma}\\&+\sum_i\left\{\sum_{k=1}^{3}\varepsilon_{\psi k}X_i^{\psi k,\psi k}+\sqrt{2}t(X_i^{\psi 1,\psi 2}+X_i^{\psi 1,\psi 3}+\text{H.c.})\right\}\\&+\sum_i\sum_m\varepsilon_\tau X_i^{\tau m,\tau m}+\sum_i\left\{\sum_{k=1}^{3}\varepsilon_{\tilde\psi k}X_i^{\tilde\psi k,\tilde\psi k}\right.\\&\left.+t(X_i^{\tilde\psi 1,\tilde\psi 2}+\text{H.c.})\right\}\\&+\sum_i\sum_m\left\{\sum_{k=1}^{2}\varepsilon_{\tilde\tau k}X_i^{\tilde\tau km,\tilde\tau km}+t(X_i^{\tilde\tau 1m,\tilde\tau 2m}+\text{H.c.})\right\},\end{aligned}\tag{A7}$$

where $t=2t^{pd}\lambda_0>0$ and $X_i^{\nu\mu}=|i\nu\rangle\langle i\mu|$. Diagonalization of $H_{\rm loc}$ can be performed independently in each set of curly brackets in Eq. (A7) to give instead of $|i\nu\rangle$ [Eq. (A4)] the new one- and two-hole basis states $|ia\rangle$ as given by

$$\begin{pmatrix}|d\sigma\rangle\\|p\sigma\rangle\end{pmatrix}=T_1\begin{pmatrix}|f\sigma\rangle\\|g\sigma\rangle\end{pmatrix},\quad T_1=\begin{pmatrix}u&v\\-v&u\end{pmatrix}$$

$$\begin{pmatrix}u\\v\end{pmatrix}=\sqrt{\frac{1}{2}\left\{1\pm\frac{|\tilde\Delta|}{\sqrt{\tilde\Delta^2+4t^2}}\right\}},\quad \tilde\Delta=\varepsilon_p-\varepsilon_d-t^{pp}\mu_0,$$

$$\varepsilon_{f,g}=\frac{1}{2}\{(\varepsilon_d+\varepsilon_p-t^{pp}\mu_0)\mp\sqrt{\tilde\Delta^2+4t^2}\},$$

$$|\psi_i\rangle=\sum_{k=1}^{3}\beta_{ik}|c_k\rangle,\quad \beta_{ik}=\{T_2\}_{ik},\tag{A8}$$

$$T_2^{-1}\begin{pmatrix}\varepsilon_{\psi 1}&\sqrt{2}t&\sqrt{2}t\\\sqrt{2}t&\varepsilon_{\psi 2}&0\\\sqrt{2}t&0&\varepsilon_{\psi 3}\end{pmatrix}T_2=\begin{pmatrix}\varepsilon_{c1}&0&0\\0&\varepsilon_{c2}&0\\0&0&\varepsilon_{c3}\end{pmatrix},$$

$$\begin{pmatrix}|\tilde\psi_1\rangle\\|\tilde\psi_2\rangle\end{pmatrix}=T_1\begin{pmatrix}|\tilde c_1\rangle\\|\tilde c_2\rangle\end{pmatrix},\quad |\tilde\psi_3\rangle\equiv|\tilde c_3\rangle,$$

$$\varepsilon_{\tilde c1,\tilde c2}=\frac{1}{2}\{(\varepsilon_d+3\varepsilon_p+t^{pp}\mu_0)\mp\sqrt{\tilde\Delta^2+4t^2}\},$$

$$\varepsilon_{\tilde c3}=\varepsilon_{\tilde\psi 3},$$

$$\begin{pmatrix}|\tilde\tau_1 m\rangle\\|\tilde\tau_2 m\rangle\end{pmatrix}=T_1\begin{pmatrix}|t_1 m\rangle\\|t_2 m\rangle\end{pmatrix},$$

$$\varepsilon_{t1}=\varepsilon_{\tilde c1},\quad \varepsilon_{t2}=\varepsilon_{\tilde c2}.$$

The u and v components of the matrix T_1 and corresponding energies ε_a for some new states are written explicitly in Eq. (A8), while the components β_{ik} of the orthogonal 3×3 matrix T_2 and energies ε_{ck} for the singlet states $|c_k\rangle$ $(k=1,2,3)$ should be found numerically with a particular choice for the parameters of the underlying p-d model.

The zeroth-order Hamiltonian now takes the form

$$H_{\rm loc}=\sum_{i,a}\varepsilon_a X_i^{aa},$$

and for the intercell interactions $H_{\rm hop}$ we have $(i\neq j)$

$$\begin{aligned}H_{\rm hop}=&\sum_{i,j,\sigma}\sum_{a,a',b,b'}[t_{ij,\sigma}(a',a|b',b)\\&+\tilde t_{ij,\sigma}(a',a|b',b)]X_i^{a',a}X_j^{b',b},\end{aligned}$$

$$\begin{aligned}t_{ij,\sigma}(a',a|b',b)=&2t^{pd}\lambda_{ij}[\langle a'|d_\sigma^\dagger|a\rangle\langle b'|p_\sigma|b\rangle+\langle a'|p_\sigma^\dagger|a\rangle\\&\times\langle b'|d_\sigma|b\rangle]-t^{pp}\mu_{ij}\langle a'|p_\sigma^\dagger|a\rangle\\&\times\langle b'|p_\sigma|b\rangle,\end{aligned}\tag{A9}$$

$$\begin{aligned}\tilde t_{ij,\sigma}(a',a|b',b)=&t^{pp}\nu_{ij}[\langle a'|p_\sigma^\dagger|a\rangle\langle b'|\tilde p_\sigma|b\rangle+\langle a'|\tilde p_\sigma^\dagger|a\rangle\\&\times\langle b'|p_\sigma|b\rangle].\end{aligned}$$

In Eq. (2.1) the terms $\tilde p_{i\sigma}^\dagger\tilde p_{j\sigma}$ with $i\neq j$ describe transitions between high-lying states and hence do not contribute to the low-energy physics. So we drop these terms in $H_{\rm hop}$ from the beginning. To derive the low-energy effective Hamiltonian $H_{\rm eff}$, the Hamiltonian $H_{\rm hop}$ should be reduced further. Below we write down explicitly the amplitudes $t_{ij,\sigma}$ and $\tilde t_{ij,\sigma}$ for the processes that contribute to $H_{\rm eff}$ up to second order.

To first order the only hopping amplitudes for the lowest singlet $|c_1\rangle$ are

$$\begin{aligned}&t_{ij,\sigma}(c_1,f\overline{\sigma}|f\overline{\sigma},c_1)\\&\quad=-2t^{pd}\lambda_{ij}[v\beta_{11}-\sqrt{2}u\beta_{31}][u\beta_{11}-\sqrt{2}v\beta_{21}]\\&\qquad-\frac{1}{2}t^{pp}\mu_{ij}[u\beta_{11}-\sqrt{2}v\beta_{21}]^2\\&\quad\equiv t_{ij}.\end{aligned}\tag{A10}$$

Below we denote this interaction as the $H_t^{(1)}$ term in $H_{\rm eff}$. There are several kinds of processes that contribute to $H_{\rm eff}$ in second order. Namely, one kind of relevant process is given by the amplitudes

$$\begin{aligned}t_{ij,\sigma}(c_k,f\overline{\sigma}|f\overline{\sigma},c_1)=&2t^{pd}\lambda_{ij}(a_kb_1+b_ka_1)-t^{pp}\mu_{ij}b_kb_1\\\equiv&F_{ij}^{ck}\quad(k=2,3),\end{aligned}$$

$$\begin{aligned}t_{ij,\sigma}(\tau m,f\sigma'|f\overline{\sigma},c_1)=&-\operatorname{sgn}(2\sigma)C^m(\sigma',\sigma)[2t^{pd}\lambda_{ij}(ua_1\\&+vb_1)-t^{pp}\mu_{ij}ub_1]\\\equiv&-\operatorname{sgn}(2\sigma)C^m(\sigma',\sigma)F_{ij}^{\tau},\end{aligned}\tag{A11}$$

$$\tilde{t}_{ij,\sigma}(\tilde{c}_1, f\overline{\sigma}|f\overline{\sigma}, c_1) = t^{pp}\nu_{ij}\frac{b_1}{\sqrt{2}} \equiv F_{ij}^{\tilde{c}1},$$

$$\tilde{t}_{ij,\sigma}(t_1 m, f\sigma'|f\overline{\sigma}, c_1) = -\mathrm{sgn}(2\sigma)C^m(\sigma',\sigma)t^{pp}\nu_{ij}b_1 \equiv -\mathrm{sgn}(2\sigma)C^m(\sigma',\sigma)F_{ij}^{t1},$$

where

$$a_k = -\frac{v}{\sqrt{2}}\beta_{1k} + u\beta_{3k} \quad , \quad b_k = \frac{u}{\sqrt{2}}\beta_{1k} - v\beta_{2k},$$

$$C^m(\sigma',\sigma) = \frac{1}{\sqrt{2}}\delta_{m,0}\delta_{\sigma',\overline{\sigma}} + \delta_{m,2\sigma}\delta_{\sigma',\sigma}. \qquad \text{(A12)}$$

Within the perturbation expansion that we develop here, these amplitudes correspond to transitions between a ground-state singlet $|jc_1\rangle$ located at the $\vec{j}$ site and an intermediate virtual upper two-hole state at a site $\vec{i}$ (the transfer to the states $|\tilde{c}_2\rangle$ and $|t_2 m\rangle$ is forbidden). Due to a discharging process of a virtual state the lowest singlet appears at another $\vec{j}'$ site.This kind of second-order hopping is presented below as the $H_t^{(2)}$ term in H_{eff}. The rest of the processes also giving rise to second-order corrections to H_{eff} have the amplitudes

$$t_{ij,\sigma}(c_k, f\overline{\sigma}|0, f\sigma) = \mathrm{sgn}(2\sigma)\left\{2t^{pd}\lambda_{ij}\left[\frac{\beta_{1k}}{\sqrt{2}} - uv(\beta_{2k} + \beta_{3k})\right] - t^{pp}\mu_{ij}\left[-\frac{uv}{\sqrt{2}}\beta_{1k} + v^2\beta_{2k}\right]\right\} \equiv \mathrm{sgn}(2\sigma)D_{ij}^{ck},$$

$$t_{ij,\sigma}(\tau m, f\sigma'|0, f\sigma) = C^m(\sigma',\sigma)[2t^{pd}\lambda_{ij}(v^2 - u^2) - t^{pp}\mu_{ij}uv] \equiv C^m(\sigma',\sigma)D_{ij}^{\tau}, \qquad \text{(A13)}$$

$$\tilde{t}_{ij,\sigma}(\tilde{c}_1, f\overline{\sigma}|0, f\sigma) = -\mathrm{sgn}(2\sigma)t^{pp}\nu_{ij}\frac{v}{\sqrt{2}} \equiv -\mathrm{sgn}(2\sigma)D_{ij}^{\tilde{c}1},$$

$$\tilde{t}_{ij,\sigma}(t_1 m, f\sigma'|0, f\sigma) = C^m(\sigma',\sigma)t^{pp}\nu_{ij}v \equiv C^m(\sigma',\sigma)D_{ij}^{t1}.$$

The corresponding processes lead to the superexchange interaction (the H_J term in H_{eff}) between the localized spin states $|if\sigma\rangle$. At the same time the amplitude in Eq. (A13) with $c_k = c_1$ provides also an additional second-order contribution to the lowest singlet hopping term $H_t^{(2)}$.

Collecting all the processes with amplitudes (A11) and (A13) into H' we may now write down the relevant part of the Hamiltonian as

$$H = H_0 + H_t^{(1)} + H'.$$

The interaction H' should be excluded by the Schrieffer-Wolff transformation to give the effective t-t'-t''-J model H_{eff} presented in the main text [Eq. (2.2]. The foregoing formulas specify the parameters of the effective Hamiltonian. Actually, the first-order hopping amplitudes t_{ij} in $H_t^{(1)}$ are given by Eq. (A10) and in the present calculations we keep only the largest ones t_1, t_2, and t_3 for the lowest singlet hopping between the first, second, and third neighbors, respectively. The resulting three-site hopping amplitudes $t_{jij'}^{N/S}$ in $H_t^{(2)}$ are given by

$$t_{jij'}^{N/S} = \xi_{c1}^{N/S}\frac{D_{ij}^{c1}D_{ij'}^{c1}}{\varepsilon_{c1} + \varepsilon_0 - 2\varepsilon_f} + \sum_{k=2,3}\xi_{ck}^{N/S}\frac{F_{ij}^{ck}F_{ij'}^{ck}}{\varepsilon_{ck} - \varepsilon_{c1}} + \xi_{\tau}^{N/S}\frac{F_{ij}^{\tau}F_{ij'}^{\tau}}{\varepsilon_{\tau} - \varepsilon_{c1}} + \xi_{\tilde{c}1}^{N/S}\frac{F_{ij}^{\tilde{c}1}F_{ij'}^{\tilde{c}1}}{\varepsilon_{\tilde{c}1} - \varepsilon_{c1}} + \xi_{t1}^{N/S}\frac{F_{ij}^{t1}F_{ij'}^{t1}}{\varepsilon_{t1} - \varepsilon_{c1}}, \qquad \text{(A14)}$$

where

$$\xi_{c1}^N = -\xi_{c2}^N = -\xi_{c3}^N = -\xi_{\tilde{c}1}^N = \frac{1}{2} \quad , \quad \xi_{\tau}^N = \xi_{t1}^N = -\frac{3}{4},$$

$$\xi_{c1}^S = \xi_{c2}^S = \xi_{c3}^S = \xi_{\tilde{c}1}^S = -1, \quad \xi_{\tau}^S = \xi_{t1}^S = \frac{1}{2}, \qquad \text{(A15)}$$

and ε_0 is the energy of the vacuum cell state. The parameters of the exchange part H_J have the familiar form[18]

$$J_{ij} = \sum_{k=1}^{3}\zeta_{ck}\frac{[D_{ij}^{ck}]^2}{\varepsilon_{ck} + \varepsilon_0 - 2\varepsilon_f} + \zeta_{\tau}\frac{[D_{ij}^{\tau}]^2}{\varepsilon_{\tau} + \varepsilon_0 - 2\varepsilon_f} + \zeta_{\tilde{c}1}\frac{[D_{ij}^{\tilde{c}1}]^2}{\varepsilon_{\tilde{c}1} + \varepsilon_0 - 2\varepsilon_f} + \zeta_{t1}\frac{[D_{ij}^{t1}]^2}{\varepsilon_{t1} + \varepsilon_0 - 2\varepsilon_f}, \qquad \text{(A16)}$$

where $\zeta_{ck} = \zeta_{\tilde{c}1} = 4$ and $\zeta_{\tau} = \zeta_{t1} = -2$. The value of Y_{ij} is given by the same formula as Eq. (A16) with the replacement $\zeta \to \zeta^Y$ and $\zeta_{ck}^Y = \zeta_{\tilde{c}1}^Y = -1$, $\zeta_{\tau}^Y = \zeta_{t1}^Y = -3/2$.

Up to now only intracell Coulomb interactions have been involved into the reduction procedure. Effects of static intercell interactions can be easily accounted for in a mean-field manner[18,19] if one assumes that in the reference background each cell carries a one-hole ground state $|f\sigma\rangle$. Then one finds the following shifts for the cell state energies:

$$\Delta\varepsilon_d = U_{pd}(2 - \phi_0)\langle n^p\rangle,$$

$$\Delta\varepsilon_p = U_{pd}(2 - \phi_0)\langle n^d\rangle + \frac{1}{2}U_p\left(\frac{1}{2} - \xi_0\right)\langle n^p\rangle,$$

$$\Delta\varepsilon_{\tilde{p}} = 2U_{pd}\langle n^d\rangle + \frac{1}{4}U_p\langle n^p\rangle, \qquad \text{(A17)}$$

$$\Delta\varepsilon_{\psi_1} = \Delta\varepsilon_{\tau} = \Delta\varepsilon_d + \Delta\varepsilon_p, \quad \Delta\varepsilon_{\psi_2} = 2\Delta\varepsilon_p,$$

$$\Delta\varepsilon_{\psi_3} = 2\Delta\varepsilon_d, \quad \Delta\varepsilon_{\tilde{\psi}1} = \Delta\varepsilon_{\tilde{\tau}1} = \Delta\varepsilon_d + \Delta\varepsilon_{\tilde{p}},$$

$$\Delta\varepsilon_{\tilde{\psi}2} = \Delta\varepsilon_{\tilde{\tau}2} = \Delta\varepsilon_p + \Delta\varepsilon_{\tilde{p}}, \quad \Delta\varepsilon_{\tilde{\psi}3} = 2\Delta\varepsilon_{\tilde{p}},$$

where $\langle n^{d,p}\rangle = \langle f\sigma|\Sigma_{\alpha}n_{\alpha}^{d,p}|f\sigma\rangle = u^2(v^2)$. By putting these Coulomb corrections into H_{loc} [Eq. (A7)] and performing

the diagonalization (A8) one finds the renormalized energies ε_a for the cell eigenstates $|a\rangle$. In particular, the corrected energies ε_f, ε_{ck}, and ε_τ lead to a renormalization of the exchange energies (A16). [Note that the last two terms in Eq. (A16) cancel each other.] A self-consistent determination of the averaged number of holes in the d and p orbitals ($\langle n^{d,p}\rangle$ and, correspondingly, u and v) is not necessary for a rough estimate of the effect of the static intercell Coulomb interaction. Any improvement should start by going beyond the mean-field character.

APPENDIX B

Here we calculate the matrix elements of the term $\hat{T}$ [Eq. (3.4)] within the variational basis (3.5),

$$T^{ab}(k)=\sum_j\ (\Psi_0|[\phi_l^a,\hat{T}]\phi_j^{b^+}|\Psi_0)e^{ik(j-l)}, \qquad \text{(B1)}$$

where a and b range from 0 to 4. The calculation is similar to that for the matrix elements of the kinetic energy $\hat{K}$ [Eq. 3.1)] and of the exchange part $\hat{H}_J$ [Eq. (2.2)] given in Ref. 25. The result may be expressed in terms of static correlation functions of Hubbard operators with noncoinciding lower indices

$$Z_a=\sum_s\ \langle X_i^{\sigma s}X_{i+a}^{s\sigma}\rangle, \qquad \text{(B2)}$$

$$D_{a,b}=\sum_{s_1,s_2}\langle X_i^{\sigma s_1}X_{i+a}^{s_1 s_2}X_{i+b}^{s_2\sigma}\rangle,$$

$$V_{a,b,c}=\sum_{s_1,s_2,s_3}\langle X_i^{\sigma s_1}X_{i+a}^{s_1 s_2}X_{i+b}^{s_2 s_3}X_{i+c}^{s_3\sigma}\rangle,$$

$$F_{a,b,c,d}=\sum_{s_1,s_2,s_3,s_4}\langle X_i^{\sigma s_1}X_{i+a}^{s_1 s_2}X_{i+b}^{s_2 s_3}X_{i+c}^{s_3 s_4}X_{i+d}^{s_4\sigma}\rangle,$$

where the angular brackets denote the average with the ground state $(\Psi_0|\ |\Psi_0)$ of the Heisenberg model. With that notation the matrix elements may be expressed as

$$T^{00}(k)=t^S\sum_{a_1,a_2}\ \overline{\delta}_{a_1,-a_2}D_{a_1,a_1+a_2}e^{ik(a_1+a_2)},$$

$$T^{n0}(k)=t^S\sum_a\ \overline{\delta}_{a,n}Z_a e^{ik(a-n)}$$

$$+t^S\sum_{a_1,a_2}\ \overline{\delta}_{a_1,-a_2}\overline{\delta}_{a_1,-n}V_{n,n+a_1,n+a_1+a_2}e^{ik(a_1+a_2)}, \qquad \text{(B3)}$$

$$T^{nm}(k)=\frac{t^S}{2}\overline{\delta}_{m,n}e^{ik(m-n)}+t^S\sum_a\ \overline{\delta}_{a,n}\overline{\delta}_{a,m}D_{a,a-m}e^{ik(a-n)}+t^S\sum_a\ \overline{\delta}_{a,-n}\overline{\delta}_{a,-m}D_{n,n+a}e^{ik(a+m)}$$

$$+t^S\overline{\delta}_{m,n}\overline{\delta}_{m,-n}\sum_{a_1,a_2}\ \overline{\delta}_{a_1,-a_2}\overline{\delta}_{a_1,-n}\delta_{a_1+a_2,m-n}D_{a_1,m-n}e^{ik(m-n)}$$

$$+t^S\sum_{a_1,a_2}\ \overline{\delta}_{a_1,-a_2}\overline{\delta}_{a_1,-n}\overline{\delta}_{a_2,m}\overline{\delta}_{a_1+a_2,m-n}F_{n,n+a_1,n+a_1+a_2,n+a_1+a_2-m}e^{ik(a_1+a_2)},$$

where n,m range from 1 to 4 and a,a_1,a_2 denote nearest neighbors. Additionally, the notation $\delta_{a_1,a_2}=1-\overline{\delta}_{a_1,a_2}$ was used.

To express the static correlation functions (B2) in terms of spin-spin correlators we use the spin rotational invariance of the ground state of H_J. The expectation values of the products of X operators with up to four factors were already calculated in Ref. 25, but now we develop a more general procedure. All the quantities (B2) can be written in the form

$$X_{12\cdots n}^{(n)}=\sum_{s_1,\ldots,s_n}\langle X_1^{\sigma s_1}X_2^{s_1 s_2}\cdots X_n^{s_n\sigma}\rangle. \qquad \text{(B4)}$$

To proceed, we collect all the spinlike X operators in one matrix

$$\mathbf{X}_j=\begin{pmatrix}X_j^{\uparrow\uparrow} & X_j^{\downarrow\uparrow}\\ X_j^{\uparrow\downarrow} & X_j^{\downarrow\downarrow}\end{pmatrix}=\begin{pmatrix}\frac{1}{2}+S_j^z & S_j^-\\ S_j^+ & \frac{1}{2}-S_j^z\end{pmatrix}. \qquad \text{(B5)}$$

Equivalently, one may write

$$\mathbf{X}_j=\frac{1}{2}\mathbf{I}+\vec{\boldsymbol{\sigma}}\vec{S}_j, \qquad \text{(B6)}$$

with the unity matrix $\mathbf{I}$ and the Pauli spin matrices $\vec{\boldsymbol{\sigma}}$. This has the advantage that we can write Eq. (B4) as the trace over a product of spin matrices

$$X_{12\cdots n}^{(n)}=\frac{1}{2}\mathrm{Tr}\left\langle\left(\frac{1}{2}\mathbf{I}+\vec{\boldsymbol{\sigma}}\vec{S}_1\right)\cdots\left(\frac{1}{2}\mathbf{I}+\vec{\boldsymbol{\sigma}}\vec{S}_n\right)\right\rangle. \qquad \text{(B7)}$$

Using the well-known algebra of spin matrices and the spin rotational symmetry (which leads to $\langle S_i^z\rangle=0$ and so on) one can simplify all the expectation values (B2). One finds, for instance,

$$X^{(4)}_{1234}=\frac{1}{16}+\frac{1}{4}\sum_{i,j\in(1,2,3,4)}^{\mathcal{P}}\langle\vec{S}_i\vec{S}_j\rangle+S^{(4)}_{1234}, \tag{B8}$$

where $\mathcal{P}$ means the sum over all possibilities to choose nonequal i and j from the set (1,2,3,4) (six terms). In the same way we obtain for a product of five X operators

$$X^{(5)}_{12345}=\frac{1}{32}+\frac{1}{8}\sum_{i,j\in(1,2,3,4,5)}^{\mathcal{P}}\langle\vec{S}_i\vec{S}_j\rangle+\sum_{i,j,k,l\in(1,2,3,4,5)}^{\mathcal{P}}S^{(4)}_{ijkl}, \tag{B9}$$

with

$$S^{(4)}_{1234}=\frac{1}{2}\mathrm{Tr}\boldsymbol{\sigma}_\alpha\boldsymbol{\sigma}_\beta\boldsymbol{\sigma}_\gamma\boldsymbol{\sigma}_\delta\langle S_1^\alpha S_2^\beta S_3^\gamma S_4^\delta\rangle. \tag{B10}$$

Here we assume that the summation over doubled indices is implied. Expression (B10) can also be written as

$$S^{(4)}_{i,i+a,i+b,i+c}=\Sigma_{abc}-\Sigma_{bac}+\Sigma_{cab}, \tag{B11}$$

with

$$\Sigma_{abc}=\langle(\vec{S}_i\cdot\vec{S}_{i+a})(\vec{S}_{i+b}\cdot\vec{S}_{i+c})\rangle. \tag{B12}$$

In the present calculation we approximate the last four-point spin correlation function in terms of two-point spin-correlations, decoupling them as in a linear spin wave theory:[29]

$$\Sigma_{abc}=S_aS_{c-b}+\frac{1}{3}S_bS_{c-a}+\frac{1}{3}S_cS_{b-a}. \tag{B13}$$

So the complete matrix $T^{ab}(k)$ can be expressed in terms of two-point spin correlations $S_a=\langle\vec{S}_i\cdot\vec{S}_{i+a}\rangle$.

APPENDIX C

In the main text it was stated that the damping of QP states at the top of the band possesses a threshold character. The threshold energy depends on the parameters t_n ($n=1,2,3$) and J, but in the simplest approximation for the form of a QP dispersion adopted below this energy is equal to $2J$.

Quite generally the Green's function (3.7) can be written as

$$G(\vec{k},\omega)=\frac{Z_k}{\omega-E(\vec{k})+i\eta}+G^{(inc)}(\vec{k},\omega), \tag{C1}$$

where a QP pole position $E(\vec{k})$ is the solution to

$$E(\vec{k})=\varepsilon(\vec{k})+\mathrm{Re}\Sigma(\vec{k},E(\vec{k})). \tag{C2}$$

Since there is no low-energy incoherent part of the hole spectrum, a QP may scatter only into lower QP states with emitting a magnon, which is given by the damping

$$\Gamma(\vec{k})=-\frac{1}{\pi}\mathrm{Im}\Sigma(\vec{k},E(\vec{k}))$$
$$=\sum_q M^2(\vec{k},\vec{q})Z_{k-q}\delta(E(\vec{k})-E(\vec{k}-\vec{q})-\omega_q). \tag{C3}$$

The numerical solutions of Eq. (C2) presented in Sec. IV are fitted well with $E(\vec{k})=\lambda_1\gamma_k^2+\lambda_2(\gamma_k^-)^2+\lambda_0$, where $\gamma_k^-=(1/2)(\cos k_x-\cos k_y)$ and $\lambda_1>\lambda_2>0$. The simplest approximation consists in neglecting a dispersion along the boundary of AFM BZ by setting $\lambda_2=0$. This is rather reminiscent of the QP dispersion law in the pure t-J model, where one obtains $\lambda_2\ll\lambda_1\simeq 2J$. Instead we write

$$E(\vec{k})=W\gamma_k^2+E_0, \tag{C4}$$

where the bandwidth W is assumed to be an arbitrary parameter that tends to increase from below, $W\leqslant 2J$, to a higher value $W>2J$. The advantage of this approximation is that the damping $\Gamma(\vec{k})$ can now be calculated analytically to the end.

According to Eq. (C4), the top of the QP band is at $\vec{k}_0=(0,0)$ or $(\pm\pi,\pm\pi)$ with $E(\vec{k}_0)-E_0=W$, $E(k_0-q)-E_0=W\gamma_q^2$, and

$$M^2(\vec{k}_0,\vec{q})=\frac{z^2t^2}{2N}\sqrt{1-\gamma_q^2}(1-\sqrt{1-\gamma_q^2})\equiv\frac{z^2t^2}{2N}\Lambda(\gamma_q). \tag{C5}$$

We introduce also the density of states function $\rho(\epsilon)=N^{-1}\Sigma_q\delta(\epsilon-\gamma_q)$ and take into account a weak dependence of the QP weight on a momentum $Z_k\simeq Z_0\simeq J/t$, as it follows from numerical computations. Hence the damping $\Gamma(\vec{k}_0)$ can now be expressed as

$$\Gamma(\vec{k}_0)=Z_0\frac{z^2t^2}{2W}\int_{-1}^{+1}d\epsilon\ \rho(\epsilon)\Lambda(\epsilon)$$
$$\times\delta\left(\sqrt{1-\epsilon^2}\left(\sqrt{1-\epsilon^2}-\frac{2J}{W}\right)\right). \tag{C6}$$

The δ function variable possesses two roots $\epsilon_{1,2}=\pm 1$ for any ratio $2J/W$ and two additional roots $\epsilon_{3,4}=\pm\sqrt{1-(2J/W)^2}$ if $2J/W<1$. Because $\Lambda(\pm 1)=0$, the first two roots do not contribute to $\Gamma(\vec{k}_0)$. A dramatic increase of $\Gamma(\vec{k}_0)$ appears, however, when W crosses the value $2J$ from below. The damping becomes

$$\Gamma(\vec{k}_0)=\frac{z^2t}{2}\left(\frac{2J}{W}\right)^2\rho\left(\sqrt{1-\left(\frac{2J}{W}\right)^2}\right)\sqrt{\frac{1-2J/W}{1+2J/W}}. \tag{C7}$$

Near the crossing, when $0<1-2J/W=\alpha\ll 1$

$$\Gamma(\vec{k}_0)\simeq z^2 t\frac{\sqrt{2}}{\pi^2}\sqrt{\alpha}\ln\frac{2}{\sqrt{\alpha}}. \tag{C8}$$

So the damping strongly increases to become of the order of t if $W>2J$. As a result, the QP states at the top of the band cannot survive anymore and become part of the incoherent spectrum.

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