Hole dynamics and photoemission in a t-J model for $SrCu_2(BO_3)_2$

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The motion of a single hole in a t-J model for the two-dimensional spin-gap compound $SrCu_2(BO_3)_2$ is investigated. The undoped Heisenberg model for this system has an exact dimer eigenstate and shows a phase transition between a dimerized and a Néel phase at a certain ratio of the magnetic couplings. We calculate the photoemission spectrum in the disordered phase using a generalized spin-polaron picture. By varying the interdimer hopping parameters we find a crossover between a narrow quasiparticle band regime known from other strongly correlated systems and free-fermion behavior. The hole motion in the Néel-ordered phase is also briefly considered.

Since the discovery of high-temperature superconductivity, doped antiferromagnets (AF) have been studied intensively. The pseudogap behavior observed in the high-T_c cuprates has stimulated great interest in systems with spin gaps. Several new one- and two-dimensional spin gap systems have been found experimentally. These materials are characterized by a disordered singlet ground state and a finite gap to all spin excitations. Some of the compounds which have two-dimensional ($2d$) character include the coupled spin ladder systems $SrCu_2O_3$,[1] CaV_2O_5,[2] $(VO_2)P_2O_7$,[3] $Cu_2(C_5H_{12}N_2)_2Cl_4$,[4] and the plaquette resonating-valence-bond system, CaV_4O_9.[5]

Recently the two-dimensional spin gap system $SrCu_2(BO_3)_2$ has been found by Kageyama *et al.*[6] It has a spin-singlet ground state with a spin gap ~ 30 K. The substance has additional interesting features, e.g., the high-field magnetization was observed to have two plateaus at 1/4 and 1/8 of the full moment. Recent work[7] suggests that the underlying physics can be understood on the basis of a two-dimensional $S=\frac{1}{2}$ Heisenberg model with antiferromagnetic nearest-neighbor (J_1, on links A) and next-nearest-neighbor (J_2, on links B) couplings on the lattice shown in Fig. 1. The nearest-neighbor bonds A define a unique singlet covering of the lattice. The Heisenberg model corresponding to Fig. 1 (with J_1, J_2) is, in fact, topologically equivalent to the model considered by Shastry and Sutherland.[8] For this model the singlet product state forms an exact eigenstate of the Hamiltonian at all couplings, and is the ground state in a region where the nearest-neighbor coupling J_1 dominates. On the other hand, for $J_1 \to 0$ the system becomes equivalent to the 2d square lattice AF (with nearest-neighbor coupling J_2) which has a Néel-ordered ground state. The Shastry-Sutherland model can be complemented by a coupling J_3 on the links C; the singlet product state is an eigenstate of this generalized model, too. For $J_2=J_3$ the total spin on each A bond is conserved, i.e., there is a macroscopic number of conserved quantities, and each eigenstate is characterized by the number and positions of triplets.

The Heisenberg model of Fig. 1 has been studied in Refs. 7 and 9–11 using exact diagonalization, Schwinger boson, and series expansion methods. For $J_2=J_3$ it can be mapped onto a spin-1 model which shows a first-order transition between the singlet state and a Néel-ordered state at $J_2/J_1=0.4296$. For $J_2 \neq J_3$ the situation is less clear. For $J_3=0$ the numerical results provide evidence for a weak first-order singlet-Néel transition at $J_2/J_1=0.70\pm0.01$. The disordered $SrCu_2(BO_3)_2$ compound lies probably close to the transition line to a Néel state, which explains the unusual temperature dependence of magnetic properties. A fit of the susceptibility obtained from the model (with J_1 and J_2 only) to the experimental data[7] leads to the estimates of $J_1=100$ K and $J_2/J_1=0.68$. However, the actual ratio of the couplings J_3/J_2 is not known.

To include the doping degree of freedom into the Shastry-Sutherland model we consider a standard t-J model on the $SrCu_2(BO_3)_2$ lattice:

$$\mathcal{H}=\sum_{(i,j)\in A}\left[-t_1(\hat{c}^\dagger_{i\sigma}\hat{c}_{j\sigma}+\text{H.c.})+J_1\mathbf{S}_i\cdot\mathbf{S}_j\right]$$
$$+\sum_{(i,k)\in B}\left[-t_2(\hat{c}^\dagger_{i\sigma}\hat{c}_{k\sigma}+\text{H.c.})+J_2\mathbf{S}_i\cdot\mathbf{S}_k\right]$$
$$+\sum_{(i,l)\in C}\left[-t_3(\hat{c}^\dagger_{i\sigma}\hat{c}_{l\sigma}+\text{H.c.})+J_3\mathbf{S}_i\cdot\mathbf{S}_l\right]. \qquad (1)$$

The electron operators $\hat{c}^\dagger_{i\sigma}$ exclude double occupancies. We have included hopping t_3 and interaction J_3 along the C bonds. If the t-J model is derived as strong-coupling limit of

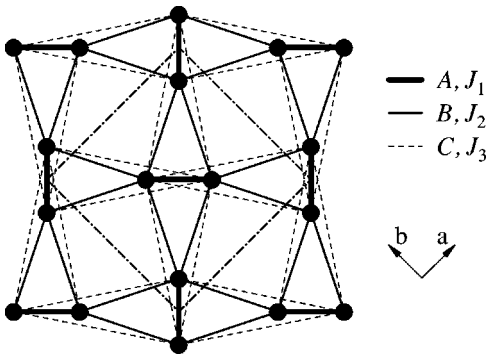

FIG. 1. Lattice structure of the Cu spins in $SrCu_2(BO_3)_2$, with the three different exchange couplings and the crystallographic axes a,b. The dash-dotted lines denote a unit cell.

a Hubbard model on the $SrCu_2(BO_3)_2$ lattice with on-site repulsion U, the ratio of the parameters is given by

$$\frac{t_1^2}{J_1}=\frac{t_2^2}{J_2}=\frac{t_3^2}{J_3}=\frac{U}{4}. \tag{2}$$

Although the compound $SrCu_2(BO_3)_2$ has, to our knowledge, not been doped so far, the study of the hole dynamics in this environment is an interesting and challenging question. Furthermore, it may be possible that finite doping leads to the formation of hole pairs and eventually to superconductivity. In this paper we shall discuss the dynamics of a single hole in an otherwise half filled system using a generalized spin-polaron picture. The one-hole spectral function for this case corresponds directly to the result of an angle-resolved photoemission experiment on the undoped compound which may give important information on the electronic correlations and the exchange constants in $SrCu_2(BO_3)_2$. We will show that the ratio t_3/t_2 tunes a crossover between a narrow-band quasiparticle (QP) behavior and a regime where a free-fermion peak dominates the spectrum. The narrow-band behavior found here is similar to other $2d$ AF systems,[12] it originates from the motion of a hole dressed with spin fluctuations.[12–15]

To investigate the hole motion we consider a one-particle Green's function describing the creation of a single hole with momentum $\mathbf{k}$ at zero temperature:

$$G(\mathbf{k},\omega)=\sum_\sigma \left\langle \psi_0^N \middle| \hat{c}^\dagger_{\mathbf{k}\sigma}\frac{1}{z-H}\hat{c}_{\mathbf{k}\sigma} \middle| \psi_0^N \right\rangle, \tag{3}$$

where z is the complex frequency variable, $z=\omega+i\eta$, $\eta\to 0$. $|\psi_0^N\rangle$ is the ground state of undoped system, i.e., with N electrons on N lattice sites.

To describe the dimerized phase we employ a bond operator representation[16,17] for the spins on the nearest-neighbor bonds. For each A bond containing two $S=1/2$ spins we introduce bosonic operators for creation of a singlet and three triplet states out of the vacuum $|0\rangle$: $s^\dagger|0\rangle=(1/\sqrt{2})(|\uparrow\downarrow\rangle-|\downarrow\uparrow\rangle)$, $t_x^\dagger|0\rangle=(-1/\sqrt{2})(|\uparrow\uparrow\rangle-|\downarrow\downarrow\rangle)$, $t_y^\dagger|0\rangle=(i/\sqrt{2})(|\uparrow\uparrow\rangle+|\downarrow\downarrow\rangle)$, $t_z^\dagger|0\rangle=(1/\sqrt{2})(|\uparrow\downarrow\rangle+|\downarrow\uparrow\rangle)$, where the constraint $s^\dagger s+\Sigma_\alpha t_\alpha^\dagger t_\alpha=1$ has to be imposed on each bond to restrict the possible states to the physical Hilbert space. The original spins are related to the new boson basis operators by $S^\alpha_{1,2}=\frac{1}{2}(\pm s^\dagger t_\alpha \pm t_\alpha^\dagger s - i\epsilon_{\alpha\beta\gamma}t_\beta^\dagger t_\gamma)$. The Hamiltonian of the Shastry-Sutherland model written in terms of the bond operators contains no terms which create triplet excitations from a state containing only singlets which means that the singlet product state $|\phi_0\rangle=\Pi_i s_i^\dagger|0\rangle$ is an exact eigenstate of the undoped system at all couplings.

If we remove one electron from an A bond, a single-hole state on this bond is created. We introduce fermionic operators for bonding (symmetric) and antibonding (antisymmetric) states of one electron (or hole) on an A bond:

$$a^\dagger_{s,\sigma}|0\rangle=\frac{1}{\sqrt{2}}(\hat{c}^\dagger_{1,\sigma}+\hat{c}^\dagger_{2,\sigma})|0\rangle,$$

$$a^\dagger_{a,\sigma}|0\rangle=\frac{1}{\sqrt{2}}(\hat{c}^\dagger_{1,\sigma}-\hat{c}^\dagger_{2,\sigma})|0\rangle. \tag{4}$$

The operators $\hat{c}^\dagger_{1,2,\sigma}$ create an electron with spin σ on one of the two sites of an A bond. Hopping via t_1 leads to an ''on-bond'' energy of $\pm t_1$ for a_a and a_s, respectively. The other hopping terms in $\mathcal{H}$ permit the hole to hop between bonds; the exchange interaction gives rise to spin fluctuations on neighboring bonds. The full interaction Hamiltonian in terms of s, t, and a operators can be easily found by calculating all possible one-hole matrix elements of the initial Hamiltonian $\mathcal{H}$ (1) (see, e.g., Ref. 18).

The evaluation of the Green's function (3) is done using the Mori-Zwanzig projection technique. The set of dynamic variables is constructed from generalized path operators[13,19] which here create strings of triplet excitations attached to the hole. Details of the calculational procedure can be found in Refs. 19 and 20. In the disordered phase of the Shastry-Sutherland model the evaluation of the matrix elements is simplified by the fact that the undoped ground state does not contain background spin fluctuations, i.e., no cumulant expectation values are involved (cf. Ref. 20). In the present calculations we have employed up to 1800 dynamic variables with a maximum path length of 3. The neglect of the self-energy terms leads to a discrete set of poles for the Green's functions, so the present approach cannot account for linewidths. In all figures we have introduced an artificial linewidth of $0.2t_1$ to plot the spectra.

Before turning to the hole dynamics it is worth mentioning that even a static vacancy (equivalent to the limit $t\to 0$) has nontrivial consequences: The pure singlet state with one spin removed, $\hat{c}_{i\sigma}|\phi_0\rangle$, is no longer an eigenstate of $\mathcal{H}$. The unpaired spin leads to triplet excitations in its neighborhood (''screening cloud''), these are spatially confined as long as the spin gap is nonzero.

Now we consider the case of nonzero hopping. We start with the ''symmetric'' choice $t_2=t_3$, $J_2=J_3$, which allows us to obtain several results analytically. $J_2=J_3$ implies that triplets are strictly localized in the absence of the hole, so any triplet created by hole hopping remains on its bond until the hole returns. $t_2=t_3$ reduces the number of possible inter-bond hopping processes. The only nonzero hopping matrix elements are $|\langle a_a s|\mathcal{H}|s a_a\rangle|=|\langle a_s t_\alpha|\mathcal{H}|t_\alpha a_s\rangle|=|\langle a_s s|\mathcal{H}|t_\alpha a_a\rangle|=t_2$ where $|XY\rangle=X_i^\dagger Y_j^\dagger|0\rangle$ is a shorthand notation for a state of two neighboring bonds i and j. It follows that a hole in the antisymmetric state a_a can freely propagate in a singlet background without emission of triplet fluctuations (direct hopping), i.e., $|\psi_{a,\mathbf{k}}\rangle=\Sigma_i \exp(i\mathbf{k}\mathbf{R}_i)a^\dagger_{a,i}s_i|\phi_0\rangle$ is an exact eigenstate of $\mathcal{H}$. In contrast, a hole being in the symmetric state a_s always creates a triplet (and converts into a_a) when it hops to a neighboring singlet bond. This in turns means that any one-hole eigenstate of $\mathcal{H}$ which contains components with a_s also involves triplet excitations. Since the triplets are localized and can only be created/removed by the hole, the state itself is localized, and contributions from such eigenstates to the spectrum are momentum-independent (nondispersive).

The calculated spectrum [Fig. 2(a)] is therefore easily understood: It shows two dispersing bands which correspond to hole hopping in the antisymmetric state a_a with effective hopping amplitude $t_2=t_3$ through the square lattice of rungs (two bands arise from the fact that the unit cell contains two rungs). All other contributions are localized and involve

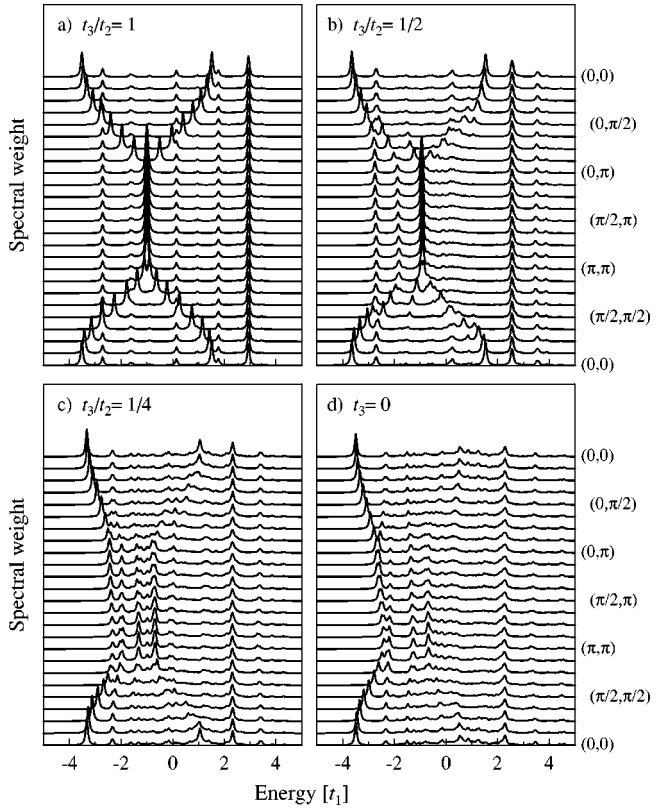

FIG. 2. Evolution of the one-hole spectral function $-\mathrm{Im}\,G(\mathbf{k},\omega)$ under a change of the hopping ratio t_3/t_2. (a) ''Symmetric'' case $t_3/t_2=1$, $J_2/J_1=0.4$; (b) $t_3/t_2=1/2$, $J_2/J_1=0.5$, (c) $t_3/t_2=1/4$, $J_2/J_1=0.6$; (d) $t_3=0$, $J_2/J_1=0.68$. The other parameters are chosen according to Eq. (2) with $U/t_1=4$. The energies are measured in units of t_1 relative to the energy of a localized hole. The ratio of the magnetic couplings is chosen to place the system close to the boundary of the singlet phase (Refs. 7 and 11), i.e., the spin gap in units of J_1 is nearly equal in the four cases.

symmetric hole states. The corresponding wave functions can be modeled by a particle moving in an attractive potential centered at a single site i. A reasonable approximation for the states contributing to the spectrum is $(ua^\dagger_{s,i}s_i + \Sigma_{\mathbf{R},\alpha} v_{\mathbf{R}} a^\dagger_{d,i+\mathbf{R}} s_{i+\mathbf{R}} t^\dagger_{i\alpha} s_i)|\phi_0\rangle$ where u,v are some coefficients (typically $|u| \gg |v_{\mathbf{R}}|$ and $v_{\mathbf{R}}$ rapidly decaying with distance). Components with more than one triplet have coefficients much smaller than the one-triplet coefficients $v_{\mathbf{R}}$. Variation of the model parameters (keeping $t_2=t_3$, $J_2=J_3$) only results in small changes in the spectrum of Fig. 2(a) since the dominating bands are determined by t_2 only. Note, however, that varying t_1/t_2 shifts the dispersing bands with respect to the localized peaks, i.e., it can induce a level crossing at momentum (0,0), so $t_1 \gg t_2$ eventually leads to a localized one-hole ground state.

Having understood the special situation at $t_2=t_3$, $J_2=J_3$, we turn to the general case. We assume $J_3<J_2$ [and $t_3<t_2$ because of Eq. (2)] since the model behavior is symmetric with respect to the interchange of the B and C bonds. In the following we discuss the t and J parameters separately, but we keep in mind that they are usually connected by the relation (2). For $J_2 \neq J_3$ the triplets are no longer completely localized, however, as is known from the undoped model, triplet hopping does not occur up to sixth order of perturbation theory in J_2, and so the triplet dispersion is weak. Consequently, $J_2 \neq J_3$ introduces a weak dispersion into the ''background features'' of the spectrum. The effect of $t_2 \neq t_3$ on the spectrum is more pronounced: Hopping of the antisymmetric hole state now also emits triplets, i.e., $|\psi_{a,\mathbf{k}}\rangle$ is no longer an exact eigenstate of $\mathcal{H}$. Therefore the well-defined dispersing bands of Fig. 2(a) mix with the background features. Spectral weight is transferred to the bottom of the spectrum which is easily understood from the fact that any triplet excitation increases the energy by at least the gap size. Figures 2(a)–(d) show the evolution of the spectrum when going from $t_3/t_2=1$ to $t_3/t_2=0$; we have fixed $U/t_1=4$ and chosen the remaining parameters such that the size of the spin gap is (approximately) preserved. For $t_3=J_3=0$, Fig. 2(d), we arrive at a situation with a narrow band at the bottom of the spectrum with dispersion minimum at (0,0); this band corresponds to the motion of a hole surrounded by triplet fluctuations (spin polaron). At higher energies a weak background is visible, it arises from excited polaron states. In addition, we note that smaller relative hopping strengths t/J suppress triplet fluctuations and reduce the high-energy background, whereas larger values of t/J lead to an incoherent spectrum since the size of the spin polaron increases.

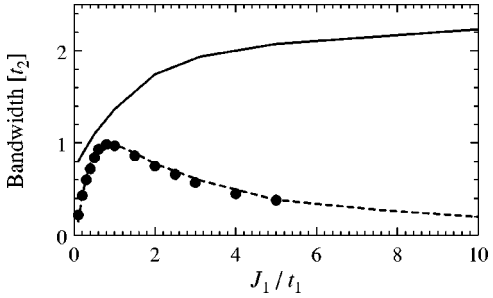

FIG. 3. Comparison of hole QP bandwidths in units of the (interdimer) in-plane hopping amplitude. Solid: $SrCu_2(BO_3)_2$ lattice, $t_2/t_1=0.82$, $J_2/J_1=0.68$, $t_3=J_3=0$, present calculation. Dashed: Square lattice, obtained by the methods used in this paper. Circles: Square lattice SCBA results from Ref. 14.

To make contact with possible experiments we examine briefly the properties of the pronounced bands at the lower edge of the spectrum which would be visible in a photoemission spectrum. The experimentally measured bandwidth can be used to determine the ratio t/J, or equivalently, the on-site repulsion U. For the (experimentally unrealistic) case of $t_2=t_3$, $J_2=J_3$, two bands should be observed as in Fig. 2(a). The width of each of the bands is given by $4t_2$ since the dispersion is the one of a free fermion. More likely, the material has $t_3 \ll t_2$, $J_3 \ll J_2$, which corresponds to Fig. 2(c) or (d). As a guide we plot in Fig. 3 the bandwidth for $t_3=J_3=0$ as function of J_1/t_1. In contrary to the well-known square-lattice case (shown for comparison) the bandwidth is finite for $J \to 0$. The reason is the possibility of direct hopping (without emission of spin excitations); this is not the case in a square lattice antiferromagnetic background where hopping always creates spin defects which have to be removed by exchange processes. For $t/J \to 0$ the bandwidth saturates (in units of t_2) at a nontrivial value which arises from the finite cloud of triplet excitations around a static hole. (Of course, for $J_2=J_3=0$ this effect is absent, and the bandwidth becomes $2t_2$ in the limit of $t/J \to 0$.)

We have also calculated the spin correlations near the mobile hole. Except for the case corresponding to Fig. 2a which shows no interdimer correlations in the ground state, the hole always introduces antiferromagnetic correlations between different dimers in its vicinity. However, for the possibly relevant values ($J_2/J_1=0.68$, $U/t_1\sim 2-10$) the dimerization is strong and the polaron size can be estimated to be smaller than three lattice spacings.

We briefly mention that the present analysis can be extended to the Néel-ordered phase of the Shastry-Sutherland model as well. Therefore a condensate of one type of triplets is introduced by a proper transformation of the basis states on each bond;[20] the condensation amplitude can be extracted from series expansion results.[10] The ground state of the undoped system is obtained by an expansion around a modified product state (which is the Néel state in the case of dominating J_2); here of course the ground state contains fluctuations around the product state. The spin polaron consists of spin deviations from the undoped background state in the vicinity of the hole. The results show that with increasing antiferromagnetic correlations the band minimum is shifted from (0,0) to (π,π) [this is equivalent to $(\pi/2,\pi/2)$ in the Brillouin zone of the square lattice defined by the B bonds]; the bands approach the shape known from the square-lattice antiferromagnet.

Summarizing, in this paper we have studied the one-hole dynamics in a t-J model for the $2d$ spin gap material $SrCu_2(BO_3)_2$. Using a generalized spin-polaron concept together with an expansion around a singlet product state we have calculated the one-hole spectral function. It shows an interesting crossover from free-fermion behavior to correlated behavior under a variation of the ratio t_2/t_3 of second and third-nearest-neighbor coupling. At $t_3=t_2$ the main contributions to the spectrum can be described by the hopping of the bare hole between the singlet rungs with a tight-binding dispersion. In contrast, for $t_3\ll t_2$ one finds a narrow band at the bottom of the spectrum and a background at higher energies; this structure can be attributed to the motion of a hole dressed with spin fluctuations (similar to other strongly correlated systems). We add that very large t/J leads to one-hole ground states with higher spin and ferromagnetic correlations in spirit of the Nagaoka effect, this has not been considered here. The hole dynamics at finite doping and the possibility of hole pairing are interesting subjects of future research.

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