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Comparison of 32-site exact-diagonalization results and ARPES spectral functions for the antiferromagnetic insulator $Sr_2CuO_2Cl_2$

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We explore the success of various versions of the one-band t-J model in explaining the full spectral functions found in angle-resolved photoemission spectra for the prototypical, quasi-two-dimensional, tetragonal, antiferromagnetic insulator $Sr_2CuO_2Cl_2$. After presenting arguments justifying our extraction of $A(\mathbf{k},\omega)$ from the experimental data, we rely on exact-diagonalization results from studies of a square 32-site lattice, the largest cluster for which such information is presently available, to perform this comparison. Our work leads us to believe that (i) a one-band model that includes hopping out to third-nearest neighbors, as well three-site, spin-dependent hopping, can indeed explain not only the dispersion relation, but also the quasiparticle lifetimes—only in the neighborhood of $\mathbf{k}=(\pi/2,0)$ do we find disagreement; (ii) an energy-dependent broadening function, $\Gamma(E)=\Gamma_0+AE$, is important in accounting for the incoherent contributions to the spectral functions. [S0163-1829(97)07034-3]

I. INTRODUCTION

The study of strongly correlated electronic systems in two dimensions (2D) has been very active, particularly since the discovery of the high-T_c superconductors. However, while there have been great improvements in the reliability and scope of experimental measurements, this has not led to agreement as to the correct description of either high-temperature superconductivity, in particular, or two-dimensional correlated systems, in general. Quite the opposite is, in fact, apparent—certain experimental results are consistent with one phenomenology, but not all experiments seem to support the same physical concepts, and as of yet no one theory is capable of explaining all the data. One avenue that may alleviate this confusion and allow for a consensus to develop follows from the study of *ideal experimental systems*. By this we imply certain compounds are amenable, for various physical as well as circumstantial reasons, to a complete characterization. Further, these compounds are such that a relatively simple theoretical model, believed to be appropriate for that material, can be defined, and with suitable ingenuity we can expect to carry out a fully quantitative comparison of this model and the experimental data.

A potential paradigm of 2D antiferromagnetic (AFM) insulators, a category which includes the parent compounds of high-T_c superconductors, is the $Sr_2CuO_2Cl_2$ compound.[1] The features that make this compound ideal are (i) it remains tetragonal down to 10 K; (ii) it has an extremely weak coupling between CuO_2 planes, and is thus a more exaggerated quasi-2D system than most high-T_c systems; and (iii) this material seems to be extremely difficult to dope away from half filling using normal preparation techniques (a summary of this behavior may be found in Ref. 1). Further, at present our experimental understanding of this material is nearly complete. Its structure has been determined by x-ray and neutron diffraction[2,3] to be body-centered-tetragonal (K_2NiF_4 type), and no transition to the orthorhombic phase is observed down to at least 10 K. This compound has a three-dimensional AFM structure and a Néel temperature $T_N=251\pm5$ K. In fact, this compound is considered to be the best available experimental realization of the $S=\frac{1}{2}$ 2D square lattice Heisenberg antiferromagnet (2DSLHA),[3,4] with nearest-neighbor exchange $J=125\pm6$ meV. Lastly, resistivity measurements show that it is strongly insulating.[2]

The (essentially) undopable character of this compound makes it an ideal candidate for angle-resolved photoemission (ARPES) experiments. Such studies allow for the measurement of the spectral function $A^{(-)}(\mathbf{k},\omega)$ [simply abbreviated as $A(\mathbf{k},\omega)$ from now on] of a *single hole* (left behind by the creation of a photoelectron) propagating in a CuO_2 plane, and such data has recently become available.[5] The results from this experiment thus provide an ideal testing ground for theories that purport to mimic the low-energy physics of CuO_2 planes; *e.g.*, the most common model, the so-called t-J model, describes spinless vacancies hopping between neighboring sites in a $S=\frac{1}{2}$ 2DSLHA. If this model is indeed a good representation of a single carrier moving in this plane, it should reproduce the *full spectral functions* measured in Ref. 5. The point of this paper is to demonstrate how well such models of carrier motion in strongly correlated systems represent the spectral properties via a comparison with the full, measured spectral functions of Ref. 5.

A. Theoretical comparisons

Not surprisingly, the ARPES results of Ref. 5 have been the subject of many theoretical studies. For example, it has

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been noted that the single-hole t-J model cannot reproduce the experimental band structure everywhere in the Brillouin zone, particularly along the (0,0) to (π,0), and the (π,0) to (0,π) directions.[5] Instead, at the very least hoppings beyond near neighbor must be added to the t-J model.[6–10] The recognition of the importance of further hoppings has also been obtained in a spin-density-wave treatment of the one-band Hubbard model.[11] Work on the more physically plausible three-band model has shown that this model is superior in reproducing the experimental band structure in all regions of the first Brillouin zone.[7,12,13] However, three-band models may be unnecessarily complicated, and thus efforts to find a useful and quantitatively accurate one-band model are warranted.

The above comparisons were based on theoretically determined and experimentally inferred dispersion relations. Of course, experimentally much more information is available, namely, the entire spectral function is known at many wave vectors. Further, it would be best if the comparison of theory and experiment was based on exact theoretical results. This is not possible unless one treats the strong-coupling Hamiltonians numerically. Recently we completed an exact-diagonalization study on a 32-site square lattice for the t-J model,[14] a formidable task (computationally speaking). This achievement is significant because exact-diagonalization results obtained with the smaller 4×4 fully square cluster are subject to the oddity that this lattice is equivalent to the four-dimensional hypercube (in the absence of interactions longer than nearest neighbor). Other exact results have been obtained on smaller, nonsquare clusters, or on clusters that lack important wave vectors (namely, those along the AFM Brillouin-zone boundary); a summary of many of these results is contained in a recent review.[15] And lastly and perhaps most importantly, while analytical work based on the self-consistent Born approximation for the t-J model has been shown to be very reliable, at least as far as the dispersion relations are concerned,[14] so far no analogous demonstration of the accuracy of this approach to include further-than-nearest-neighbor hoppings of the t-J model has been published. Thus, we consider our numerical, unbiased, 32-site exact-diagonalization approach as a precise manner in which these models can be compared to experiment.

In the present paper we continue our numerical studies on the 32-site square lattice, and present a comparison of the full spectral functions for the so-called t-t'-t''-J model, a generalization of the simpler t-J model that includes further hopping processes, as well as three-site, spin-dependent hopping. Various groups have claimed that this model accurately fits the dispersion relations found in the AFM insulator $Sr_2CuO_2Cl_2$, and such claims are something that we can critique with our numerical results. Here, besides continuing these comparisons with our numerical work, we propose an energy dependence of the self-energy for the propagating spin polarons.

B. Experimental analysis

Angle-resolved photoelectron spectroscopy (ARPES) has proven to be an invaluable experimental tool for the understanding of the electronic structure of the cuprate superconductors and related correlated-electron materials. The analysis of the ARPES results depends on the emission process being primarily direct transitions so that the spectra reflect the spectral function $A(\mathbf{k},\omega)$. This has allowed for the extensive use of ARPES to determine energy versus $\mathbf{k}$ relations for the highest energy band in the cuprates, and particularly for determining the Fermi surfaces of the metals. Most comparisons between calculations and experiment have been performed by extracting a quasiparticle energy at each $\mathbf{k}$ from both the experimental spectra and the calculated $A(\mathbf{k},\omega)$ and comparing the dispersions thus derived. However, since both experiment and theoretical calculations represent the spectral function, it should be more instructive to compare the two directly rather than just comparing the derived dispersions. A direct comparison of spectral functions has its own difficulties, as are outlined below, but hopefully will allow a more complete evaluation of the spectral function derived from the Green's function and thus avoid the possible errors involved in determining the experimental quasiparticle dispersion.

For two-dimensional materials such as the cuprates, the inherent line shape of the ARPES spectrum may reveal information about the nature of the elementary excitations. Many previous ARPES studies have examined the line shape of the spectral functions. (For a review of this subject, see Ref. 16; for particularly relevant experimental work, see Ref. 17.) In these studies the assumption is made that direct transitions dominate the spectral intensity, and thus one may model the spectral response with a single peak, and it has been proposed that the nature of this peak should be characteristic of a Fermi liquid, a marginal-Fermi liquid, or some other microscopic model. Since these fits have all been to a single peak they represent only the coherent part of the spectrum. Most numerical calculations of $A(\mathbf{k},\omega)$ for the cuprates indicate that there is a great deal of spectral weight in the incoherent parts of the excitations. Therefore, attempting to fit the cuprate ARPES spectra with a single peak of any shape is inherently flawed. In this work we make no attempt to deduce a specific line shape, or broadening function, but merely use a few simple fitting procedures as discussed below. The essential point is that for the purposes of fitting the experimental data, a correct form for $A(\mathbf{k},\omega)$ is more important than the detailed line shape of a single peak.

The planar copper oxides are well suited for analysis since in most of these materials there is a single highest energy occupied state (band) which is well separated in energy from any other bands. Experimentally, $Sr_2CuO_2Cl_2$ has some particular advantages for the present study in addition to those mentioned above. It is easy to prepare good surfaces for photoemission since the material cleaves in a manner similar to mica. In addition, the fact that the oxygen content is fixed to the stoichiometric value gives us confidence that the actual surface under study truly represents a CuO_2 plane with a single hole per copper site. Thus, while the line shape and broadening appropriate for a particular peak in $A(\mathbf{k},\omega)$ may be more difficult to determine than for the conducting cuprates, it should be more straightforward to determine the correct Greens function for comparison.

Our paper is organized as follows. Firstly, in Sec. II we discuss the extraction of the line shapes of the spin-polaron band from the ARPES data. Then, in Sec. III we discuss the various modifications to the t-J model necessary to improve the success of this model in explaining the ARPES results. In

Sec. IV we present our numerical results and the comparison of our work to the experimental spectral functions; conclusions are provided in Sec. V.

II. EXTRACTION OF $A(\mathbf{k},\omega)$ FROM ARPES DATA

The collection of the relevant data has already been discussed in Ref. 5, and in that paper the manner in which the quasiparticle dispersion relation was obtained from the ARPES data was presented. Here we focus on the quasiparticle line shapes.

We propose that the experimental data corresponds to a direct-transition dominated $A(\mathbf{k},\omega)$ plus a background. We need to subtract this background for comparison to the calculated spectral functions, and while there is no definitive prescription for finding this background function, enough is known to provide a useful guide. The dominant sources of background should be intensity from lower energy bands, higher-order light excitations, and inelastically scattered electrons. The first two terms are easily handled: a Gaussian represents the other bands and has little effect above approximately -1.9 eV. Higher-order light excitations give a constant background that is seen in the presence of some counts at energies greater than the Fermi energy, and are thus a zero offset for the intensity axis. The crucial question is the influence of inelastic scattering at energies around -1.5 eV. A large step function or linear background would account for much of the intensity in this region and make the data appear as a single, broad peak. This sort of approach has been used in many previous attempts deducing ARPES line shapes in high-T_c materials. However, as pointed out by Claessen *et al.*,[17] the standard procedure for finding this inelastic background gives a small contribution for the energies of interest. Consistent with this, we find that when we fit the entire spectral function the necessary inelastic background term is indeed very small. However, this inelastic term is strongly **k** dependent, which is completely unphysical; therefore, we have simply set this term to zero. The background terms we are left with are a simple constant and a Gaussian, the latter accounting for the lower energy band structure. Since this band structure is dispersive, we determine the appropriate Gaussian contribution to the background at each **k** by a least-squares fit, and subtract it and the **k** independent constant term from the experimental data.

The corrected spectra have the following characteristics: (i) the peaks broaden and weaken as **k** moves away from the valence-band maximum (VBM) at $(\pi/2,\pi/2)$, (ii) the peaks at their sharpest are broad and asymmetrical, and (iii) although the quasiparticle dispersion is flat along the (0,0) to $(\pi,0)$ direction, the spectral weight is a maximum at $(\pi/2,0)$.

As an example of the application of this procedure, in Fig. 1 we show the ARPES intensities and the background function that we have employed to obtain the quasiparticle $A(\mathbf{k},\omega)$ for $\mathbf{k}=(\pi/2,\pi/2)$. Clearly, the peak arising from the main valence band is very well represented, even without the inclusion of the inelastic linear term — similar satisfactory agreement is found at other wave vectors. Thus, we hope that by following this prescription we are comparing our theoretical work to the intrinsic spectral response of a single hole propagating in a CuO$_2$ plane in Sr$_2$CuO$_2$Cl$_2$.

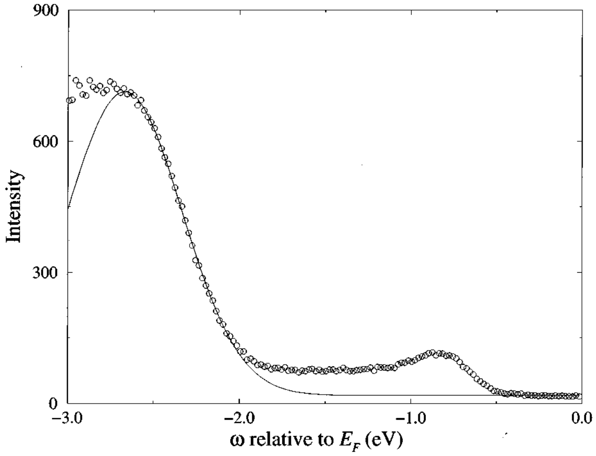

FIG. 1. The background function for the main valence band of the insulator (solid line) superimposed on the ARPES data, represented by open circles (○), for $\mathbf{k}=(\pi/2,\pi/2)$. The remaining spectral weight is associated with the intrinsic spectral response, coherent as well as incoherent, of a single hole in a 2D AFM.

III. THEORETICAL MODELS

To study the theoretical spectrum $A(\mathbf{k},\omega)$ of the planar copper oxides, we start with the simple t-J model with Hamiltonian,

$$\mathcal{H}_1=-t\sum_{\mathrm{NN},\sigma}(\tilde{c}^{\dagger}_{i,\sigma}\tilde{c}_{j,\sigma}+\mathrm{H.c.})+J\sum_{\mathrm{NN}}\left(\mathbf{S}_i\cdot\mathbf{S}_j-\frac{1}{4}n_in_j\right), \tag{1}$$

where NN means nearest neighbors.[15] It is well known that this simple, one-band model cannot reproduce all the features of the single hole dispersion relation $E(\mathbf{k})$ of Sr$_2$CuO$_2$Cl$_2$ as measured by ARPES.[5,14] While the $E(\mathbf{k})$ from the t-J model agrees with experiment along the (0,0) to (π,π) direction, two discrepancies remain: (i) along the antiferromagnetic Brillouin zone (ABZ) edge $[(\pi,0)$ to $(0,\pi)]$, the t-J model is less dispersive; and (ii) along the (0,0) to $(\pi,0)$ direction, the model is much more dispersive than the experimental result.

Various modifications to the t-J model have been suggested to make the theoretical prediction in better agreement with experiment. One suggestion follows from the finding that a three-band model works much better than the one-band model,[7,13] viz. one can mimic many features of the three-band model in the one-band model by including farther-than-nearest-neighbor hopping terms. For example, it has become customary to include[8]

$$\mathcal{H}_2=-t'\sum_{2\mathrm{NN},\sigma}(\tilde{c}^{\dagger}_{i,\sigma}\tilde{c}_{j,\sigma}+\mathrm{H.c.})-t''\sum_{3\mathrm{NN},\sigma}(\tilde{c}^{\dagger}_{i,\sigma}\tilde{c}_{j,\sigma}+\mathrm{H.c.}), \tag{2}$$

where 2NN and 3NN are the second- and third-nearest neighbors, respectively. Inclusion of $\mathcal{H}_2$ significantly lowers the anisotropy of the effective-mass tensor found in the theoretical $E(\mathbf{k})$ around $(\pi/2,\pi/2)$,[8] but discrepancy (ii) above still remains. However, recently it has been suggested that so-called three-site, spin-dependent hopping terms[18] will

TABLE I. Quasiparticle weight $Z_{\mathbf{k}}$ and energy $E(\mathbf{k})$ of the t-t'-t''-J model with $(\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3)$ and without $(\mathcal{H}_1+\mathcal{H}_2)$ the three-site, spin-dependent hopping terms, calculated on a 32-site square lattice. The parameters that we used are $t'=-0.3t$, $t''=0.2t$, and $J=0.3t$. $E(\mathbf{k})$ is in units of t and is measured relative to the half-filled ground-state energy $E_0=-11.3297t$.

	$\mathcal{H}_1+\mathcal{H}_2$		$\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3$	
k	$Z_{\mathbf{k}}$	$E(\mathbf{k})$	$Z_{\mathbf{k}}$	$E(\mathbf{k})$
(0,0)	0.0916	1.8390	0.0195	2.0668
$\left(\frac{\pi}{4},\frac{\pi}{4}\right)$	0.1820	2.0832	0.0462	2.3120
$\left(\frac{\pi}{2},\frac{\pi}{2}\right)$	0.3527	2.3868	0.3721	2.6880
$\left(\frac{3\pi}{4},\frac{3\pi}{4}\right)$	0.0749	2.0554	0.0379	2.3314
(π,π)	0.0021	1.7661	0.0000	2.0280
$\left(\pi,\frac{\pi}{2}\right)$	0.1086	1.9741	0.0736	2.2418
$(\pi,0)$	0.0287	1.7126	0.0276	1.9733
$\left(\frac{\pi}{2},0\right)$	0.2117	1.9897	0.1150	2.2202
$\left(\frac{3\pi}{4},\frac{\pi}{4}\right)$	0.0721	1.9588	0.0340	2.2556

make the theoretical $E(\mathbf{k})$ from (0,0) to $(\pi,0)$ almost dispersionless,[9,10] thus eliminating this problem. These processes are given by

$$\mathcal{H}_3=\frac{J}{4}\sum_{j,\sigma}\sum_{\delta\neq\delta'}(\tilde{c}^{\dagger}_{j+\delta,\sigma}\tilde{c}^{\dagger}_{j,-\sigma}\tilde{c}_{j,\sigma}\tilde{c}_{j+\delta',-\sigma} - \tilde{c}^{\dagger}_{j+\delta,\sigma}n_{j,-\sigma}\tilde{c}_{j+\delta',\sigma}), \tag{3}$$

where δ and δ' are the unit vectors $\pm\hat{\mathbf{x}}$ and $\pm\hat{\mathbf{y}}$.

We have investigated the effects of these terms ($\mathcal{H}_2$ and $\mathcal{H}_3$) on $E(\mathbf{k})$ of the t-J model using the same method as in Ref. 14. In particular, we calculate the quasiparticle dispersion relation $E(\mathbf{k})$ and quasiparticle weight $Z_{\mathbf{k}}$ of these models for a 32-site square lattice. The parameters we employed are $t'=0.3t$, $t''=-0.2t$, and $J=0.3t$, and our results are tabulated in Table I. [Note that $E(\mathbf{k})$ as defined in Ref. 14 is relative to the undoped ground-state energy.] As pointed out before,[14] the behavior of $Z_{\mathbf{k}}$ of the t-J model along the ABZ edge [from $(\pi,0)$ to $(0,\pi)$] does not agree with experiment. ARPES data shows that the peak intensity is the largest at $(\pi/2,\pi/2)$, whereas $Z_{\mathbf{k}}$ for this wave vector for the t-J is the smallest along this direction. From Table I it is clear that inclusion of farther-than-nearest-neighbor hopping terms eliminates this discrepancy. We also note that the three-site hopping terms $\mathcal{H}_3$ suppress $Z_{\mathbf{k}}$ at all $\mathbf{k}$ except at the VBM $(\pi/2,\pi/2)$. Although we cannot produce a quantitative comparison of $Z_{\mathbf{k}}$ with ARPES results, we will see (in Sec. IV) that this suppression of $Z_{\mathbf{k}}$ makes $A(\mathbf{k},\omega)$ agree better with the ARPES results.

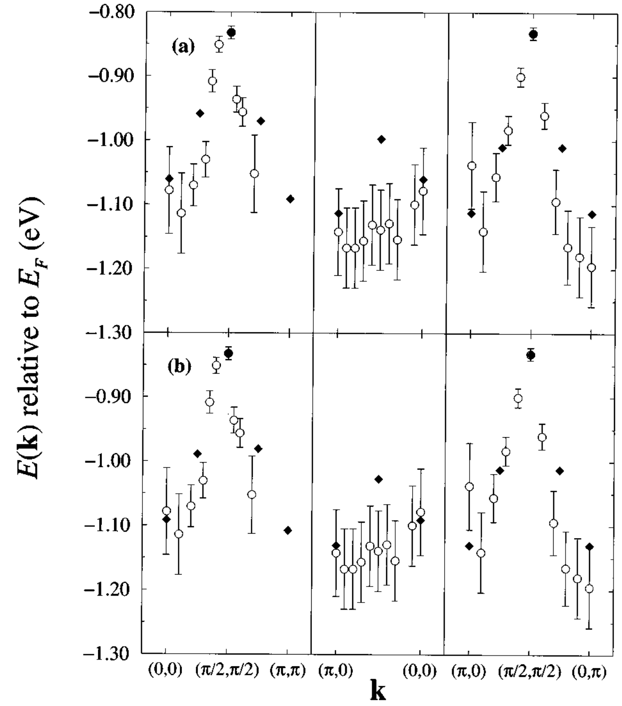

FIG. 2. Dispersion relations for single hole propagating in a 2D square lattice AFM described by the t-t'-t''-J model (a) without $(\mathcal{H}_1+\mathcal{H}_2)$ and (b) with $(\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3)$ the three-site, spin-dependent hopping processes. We have used the following parameters: $t'=-0.3t$, $t''=0.2t$, and $J=0.3t$. The open circles (○) with error bars are the ARPES results from Ref. 5.

To make a comparison of $E(\mathbf{k})$ with experimental results, we should express it in units of eV and relative to the experimental Fermi energy. This can be done by using the energy scale $J=0.125$ eV and the location of the ARPES peak at $\mathbf{k}=(\pi/2,\pi/2)$. In Fig. 2 we plot the dispersion relation of the $\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3$ with and without the three-site hopping terms, and our numerical results are compared with ARPES data.[5] Consistent with previous studies,[8] we find that adding the farther-than-nearest-neighbor hopping terms to the t-J model leads to an (essentially) isotropic effective-mass tensor near the VBM. However, there is still some dispersion along the (0,0) to $(\pi,0)$ direction, in contrast to the featureless ARPES results. Previous studies[9,10] have suggested that including the three-site hopping term $\mathcal{H}_3$ can reduce the dispersion along this direction, hence making the theoretical prediction closer to the experimental result. However, from Fig. 2(b) we see that $\mathcal{H}_3$ only lowers the energy of $(\pi/2,0)$ by a small amount (and thus our exact, unbiased, numerical results are in disagreement with earlier analytical work, as well as with work on smaller clusters). Despite the lack of perfect agreement along this direction, this is the best comparison to the dispersion relation inferred from the ARPES data for the $\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3$ Hamiltonian that we have found.

IV. CALCULATION OF $A(\mathbf{k},\omega)$ AND COMPARISON WITH ARPES

The principal quantity of interest in this paper is the electron spectral function $A(\mathbf{k},\omega)$, and is defined by

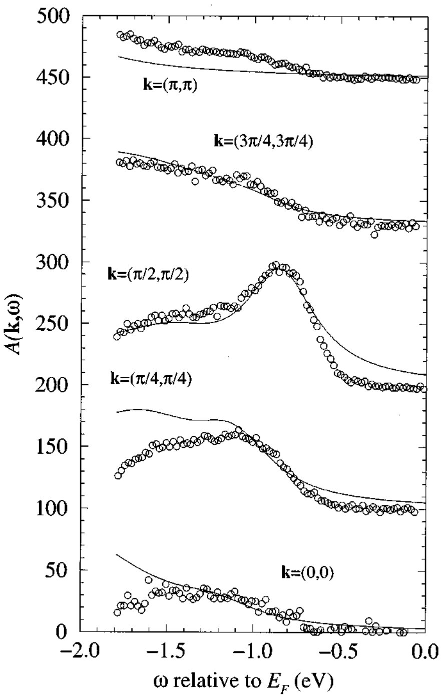

FIG. 3. Comparison of the theoretical $A(\mathbf{k},\omega)$ for $\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3$ to the ARPES data at available wave vectors **k** from (0,0) to (π,π). Solid lines are $A(\mathbf{k},\omega)$ calculated using a constant broadening $\Gamma=0.6t$ [refer to Eq. (5)] at the **k** indicated in the graph. Open circles (○) are corrected ARPES data at the same **k**. [Since the experiments were not performed at exactly the same wave vectors that we can treat numerically, for the theoretical $\mathbf{k}=(\pi/4,\pi/4)$ we show the ARPES data at $(0.3\pi,0.3\pi)$, and for the theoretical $\mathbf{k}=(3\pi/4,3\pi/4)$ we show the ARPES data at $(0.8\pi,0.8\pi)$.]

$$A(\mathbf{k},\omega)=\sum_n |\langle\psi_n^{N-1}|\tilde{c}_{\mathbf{k},\sigma}|\psi_0^N\rangle|^2\delta(\omega-E_0^N+E_n^{N-1}), \quad (4)$$

where E_0^N and ψ_0^N are the ground-state energy and wave function at half filling, respectively, and E_n^{N-1} and ψ_n^{N-1} are the energy and wave function of the nth eigenstate of the single-hole problem, respectively. To fit the line shape of the ARPES results we need to broaden the δ peaks from the theoretical calculation. The continued-fraction expansion used in Ref. 14 is equivalent to broadening the peaks by Lorentzians,

$$A(\mathbf{k},\omega)=\sum_n |\langle\psi_n^{N-1}|\tilde{c}_{\mathbf{k},\sigma}|\psi_0^N\rangle|^2 \times\frac{1}{\pi}\left[\frac{\Gamma}{(\omega-E_0^N+E_n^{N-1})^2+\Gamma^2}\right]. \quad (5)$$

The broadening factor Γ is a constant, independent of **k** and ω. To determine Γ we fit $A(\mathbf{k},\omega)$ to the ARPES at $\mathbf{k}=(\pi/2,\pi/2)$ where the peak is most well defined. For the best agreement, we have to use $\Gamma=0.6t\sim250$ meV. Note that this is as large as the coherent bandwidth, which is 280 ± 60 meV. Now, since all energy scales and Γ are fixed by the spectrum at $\mathbf{k}=(\pi/2,\pi/2)$, no fitting is needed for the other **k**.

In Fig. 3 we plot $A(\mathbf{k},\omega)$ on top of the corrected ARPES data at the available **k** from (0,0) to (π,π). (These experimental data are probably the most reliable because in the experiment the angle between the electric-field vector, the sample axes, and the ejected electrons remains constant.) Since the theoretical model is supposed to describe only the low-energy physics of the CuO_2 plane, we truncate the spectrum at $\omega=-1.8$ eV. It is obvious that the model can reproduce the experimental finding that the spectral weight is maximum at the VBM and that the peak broadens and weakens as **k** moves away from this point in the Brillouin zone. Further, we find that the incoherent part of the theoretical spectra make the peaks asymmetric, also in qualitative agreement with experiment. However, they are not as asymmetric as the experimental results, and the comparison is not entirely encouraging. Of course, the question is: is this the best this theoretical model can do?

Looking at the VBM peak, which is the sharpest and the most prominent, it is clear that we cannot completely fit the ARPES result with simple Lorentzians, since the high-energy part of the peak falls off much faster than a Lorentzian. Contrary to the ARPES of metals, this faster fall off cannot be accounted for by including a Fermi function. To improve the comparison of our numerics and experiment, without microscopic justification we use an energy-dependent broadening function $\Gamma(\omega)$. Analogous to Eq. (5), we write

$$A(\mathbf{k},\omega)=\sum_n |\langle\psi_n^{N-1}|\tilde{c}_{\mathbf{k},\sigma}|\psi_0^N\rangle|^2\frac{1}{\mathcal{N}(E_0^N-E_n^{N-1})} \times\left[\frac{\Gamma(\omega)}{(\omega-E_0^N+E_n^{N-1})^2+\Gamma^2(\omega)}\right], \quad (6)$$

where $\mathcal{N}(E)$ is the normalization factor,

$$\mathcal{N}(E)=\int_{-\infty}^{\infty}\frac{\Gamma(\omega)}{(\omega-E)^2+\Gamma^2(\omega)}d\omega. \quad (7)$$

From the spectrum at $(\pi/2,\pi/2)$ in Fig. 3, the low-energy tail (incoherent part) of the spectrum seems to need larger broadening. For simplicity we choose $\Gamma(\omega)$ to be linear in ω:

$$\Gamma(\omega)=\max(0,-0.176\ \mathrm{eV}-0.55\omega). \quad (8)$$

Note that at the first peak ($\omega\sim-0.85$ eV), $\Gamma(\omega)\sim290$ meV which is similar to the constant Γ mentioned above.

In Fig. 4 we plot $A(\mathbf{k},\omega)$ calculated from Eqs. (6) and (8) on top of the corrected ARPES data for those **k** from (0,0) to (π,π). This shows that our energy-dependent broadening function is able to produce a much improved fit at all **k**. In Figs. 5 and 6 we plot the comparison along the (0,0) to $(\pi,0)$, and the $(\pi,0)$ to $(0,\pi)$ directions, respectively. Figures 4, 5, and 6 show that our model fits the ARPES line shape at many **k** very well. The least satisfactory fit is at $\mathbf{k}=(\pi/2,0)$, which is also the least satisfactory **k** point in the dispersion relation comparison, as seen in Fig. 2. Despite this, our model does produce the largest spectral weight (cf. Table I) along the (0,0) to $(\pi,0)$ direction at this wave vector, similar to experiment.

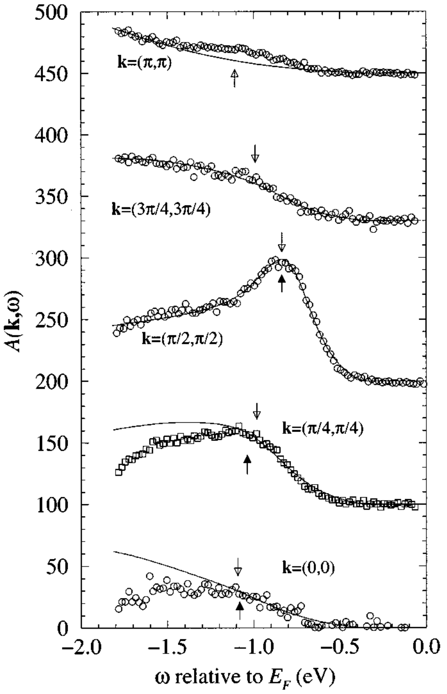

FIG. 4. This figure shows the comparison of our spectral functions to wave vectors along (k,k), the same as in Fig. 3, except that $A(\mathbf{k},\omega)$ is calculated using $\Gamma(\omega)=\max(0,-0.176\ \mathrm{eV}-0.55\omega)$. We have also included solid arrows to indicate the positions of the first peaks of the ARPES data as shown in Fig. 2, and hollow arrows to indicate the numerically determined $E(\mathbf{k})$.

In principle, we can allow the broadening function to be dependent on $\mathbf{k}$, $\Gamma(\mathbf{k},\omega)$.[17] For example, instead of determining the two coefficients in Eq. (8) using the spectrum at $(\pi/2,\pi/2)$, we can determine them at each $\mathbf{k}$. We have completed such fittings, but we do not find much better results, and in particular, the discrepancy between theoretical and experimental spectra at $(\pi/2,0)$ remains.

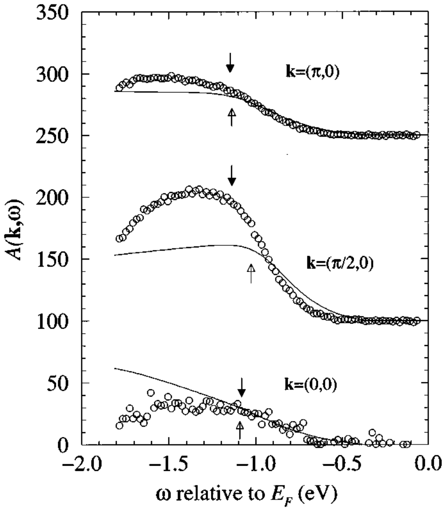

FIG. 5. Same as Fig. 4 but along the (0,0) to $(\pi,0)$ direction.

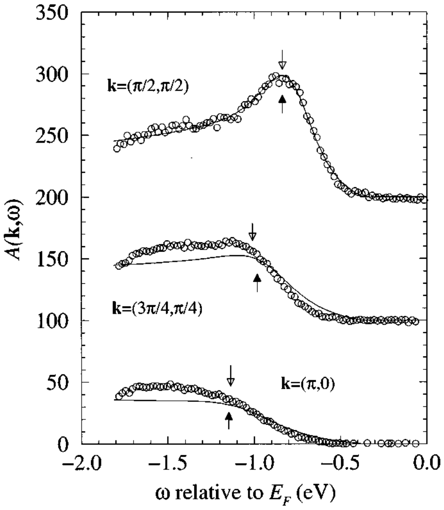

FIG. 6. Same as Fig. 4 but along the $(\pi,0)$ to $(0,\pi)$ direction. [Similar to the circumstances discussed in Fig. 3, for the theoretical $\mathbf{k}=(3\pi/4,\pi/4)$ we show the ARPES results for $\mathbf{k}=(0.3\pi,0.7\pi)$, the latter found from an average of the data for $(0.7\pi,0.3\pi)$ and $(0.3\pi,0.7\pi)$.]

V. CONCLUSIONS

To summarize, to compare the theoretical and experimental line shapes we have argued that the ARPES data of $Sr_2CuO_2Cl_2$ is dominated by direct transitions, and thus with the valence-band contribution appropriately subtracted off, one can obtain the spectral function for a single hole propagating in a CuO_2 plane. From these experimental spectral functions it is obvious that the spectrum at each $\mathbf{k}$ cannot be fit by a single, narrow peak; instead, the incoherent part is important.

We have studied the effects of including farther-than-nearest-neighbor and three-site, spin-dependent hopping terms to the t-J model. Consistent with previous studies, we find that these terms increase the dispersion along the antiferromagnetic Brillouin-zone edge [$(\pi,0)$ to $(0,\pi)$], and suppress that along the $(\pi,0)$ to (0,0) direction. In addition, these terms suppress the quasiparticle weight except at the valence-band maximum $(\pi/2,\pi/2)$. This is shown to be essential in fitting the theoretical line shape to the experimental results.

We also find that if we use an energy-dependent broadening function $\Gamma(\omega)$, we can obtain a reasonably good fit in an energy range of up to 1.8 eV below the Fermi level. The fact that we find a damping linear in energy should not be seen as direct support for the marginal Fermi-liquid hypothesis, since we are studying an insulator and thus we cannot predict that this linear term survives in the (anomalous) metallic state.

The least satisfactory fit is found along the $(\pi,0)$ to (0,0) direction, and at $(\pi/2,0)$ our calculated spectrum is significantly lower than the ARPES result. To date, only three-band models have provided a reasonable agreement with the dispersion near this point.

An interesting question that follows from our results is: What is the origin of the incoherent processes that contribute so strongly to spectral functions of a single hole propagating in a strongly correlated, half-filled state? Two possibilities seem worthy of further study. One, long-wavelength spin excitations, excluded from our work due to the use of a finite cluster, are important. Two, some other excitations, e.g., lattice waves, are strongly coupled to either the hole motion or the spin waves, or possibly both, and provide an effective energy-dependent damping of these quasiparticles.

Note added in proof. Pothuizen *et al.*[19] recently published results suggesting the existence of a $\vec{k}$-dependent broadening, similar in spirit to the ideas contained in this paper.

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