Spectral Function Evolution in a Multiband d-Electron Model: *Combined* Single-Particle and Many-Body Effects

Changfeng Chen
Department of Physics, University of Nevada, Las Vegas, Nevada 89154
(Received 1 June 1994)

We report the results of a theoretical study on the evolution of the photoemission spectral function driven by interactions in a realistic d-electron model with d^1 configuration. These results are qualitatively different from those of single-particle and simple Hubbard-type models. Both the realistic band-structure aspects and the on-site interactions are considered. Physical insights are gained about general spectroscopic behavior of multiband d-electron materials with degenerate occupied orbitals. Excellent agreement with experiment is found.

PACS numbers: 75.10.Lp, 71.45.Gm, 79.60.–i

The discovery and subsequent studies of novel materials such as high-temperature superconducting oxides, heavy-fermion compounds, and low-dimensional magnetic systems have presented a serious challenge to the understanding of the properties of strongly correlated electron systems. While these materials exhibit a wide range of extremely rich physical phenomena involving complicated physical and chemical effects, a lot of effort in this field has been devoted to the study of relatively "simple" models such as the Heisenberg model, the lattice Anderson model, the Hubbard model, and their variants with the hope of capturing fundamental physics underlying the observed new phenomena. Indeed, new concepts have been developed and interesting results obtained [1]. Because of the complexity of the materials involved, it has often become necessary in theoretical studies to incorporate various aspects of the problem by using different techniques. For example, band-structure calculations may provide the real material aspects of the system, such as the tight-binding Hamiltonian parameters for both single-particle and interaction terms, to be used as the input in *simplified* models [2–6]. Many-body techniques are then applied to study correlation effects [1]. These techniques may involve various approximations on the interaction terms [7]. On the other hand, there is another important aspect of the "realistic" description of strongly correlated systems, i.e., the suitability of the model itself. Sometimes it is necessary to include the orbital degeneracy and other band-structure aspects in the model to correctly describe qualitatively different physical phenomena. This is particularly true in the study of various spectroscopic behavior of strongly correlated systems. To interpret the observed spectra and extract correct physics, proper inclusion of some realistic band aspects are of crucial importance since some "unexpected" features may result from *combined* single-particle and many-body effects.

In this paper, we report the results of a theoretical study on the spectroscopic behavior of a realistic multiband d-electron model. This work was motivated by recent experimental studies of the evolution of the spectral function in some Mott-Hubbard systems with d^1 configuration by Fujimori *et al.* [8]. They discovered that when the ratio of the effective on-site Coulomb interaction to the single-particle bandwidth is experimentally tuned up, there is a drastic spectral weight transfer from the "coherent" part (predicted by single-particle theory) to the "incoherent" part (beyond the coherent part) in the spectra of several correlated itinerant systems. This behavior cannot be explained by either single-particle theories or simple Hubbard-model calculations. It presents a textbook quality example where both single-particle and many-body aspects must be considered on an equal footing in the theoretical modeling and calculations. In the reported experimental work [8], several material systems with different crystal and band structures are used. Here we do not intend to include all the details of the band structures in our model. Rather, we construct a *generic* d-electron model to capture the fundamental physics involved. We study the effects of many-body correlations on the spectral behavior in a model system with realistic single-particle band structures. This model is simple enough to render the many-body problem fully tractable; yet both single-particle and many-body aspects of the problem are properly considered, allowing the extraction of interesting physics that is relevant for real material systems. The calculated results have revealed some "unusual" behavior in spectral function evolution, which provides a natural explanation for the experimental results [8].

The model Hamiltonian contains both single-particle and interaction terms:

$$H = \sum_{i,j;\mu,\nu;\sigma} t_{i\mu,j\nu} c^{\dagger}_{i\mu\sigma} c_{j\nu\sigma} + \sum_{i;\mu;\sigma} E_{\mu} c^{\dagger}_{i\mu\sigma} c_{i\mu\sigma} + \sum_{i;\mu,\nu,\lambda,\phi;\sigma,\sigma'} V^{dd}_{\mu\nu\lambda\phi} c^{\dagger}_{i\mu\sigma} c^{\dagger}_{i\nu\sigma'} c_{i\lambda\sigma'} c_{i\phi\sigma} , \quad (1)$$

where the first two terms describe the hopping and orbital energies and the third term is the on-site d-d interaction which is the dominant contribution compared to intersite interactions [9]. The indices i, j label the lattice sites; μ, ν, λ, and ϕ label the d orbitals; and σ, σ' are spin

 0031-9007/94/73(14)/1982(4)$06.00

labels. All notations are standard. In this model we retain three degenerate t_{2g} d orbitals on each site. The e_g orbitals are assumed to be empty, corresponding to the experimental situation [8]. The single-particle terms are parametrized according to the Slater-Koster tight-binding scheme [10]. Only d orbitals and the nearest-neighbor hopping are explicitly included. The effects of other orbitals as well as beyond the nearest neighbors will renormalize the on-site d-electron energies [11,12]. To treat the single-particle and many-body effects on an equal footing, we employ a numerical exact diagonalization approach. We choose a tetrahedron cluster and apply periodic boundary conditions in all three dimensions to construct a bulk fcc structure [13]. This is equivalent to sampling four points (corresponding to the four sites in the tetrahedron cluster in real space) in the Brillouin zone: the Γ point and the three X points. All many-body states are generated and projected according to the symmetries inherent in the Hamiltonian, which include the total particle number in the cluster, total spin and its z component, and the spatial translational symmetry. The spatial symmetry is implemented by projecting states according to the irreducible representations of the space group of the system [11]. The symmetrized basis functions are then used to construct the Hamiltonian matrices which are in block-diagonalized form, partitioned according to various symmetry indices. Direct diagonalization of these matrices provides exact solutions to the Hamiltonian.

The single-particle energy levels in this cluster model are expressed in terms of the Slater-Koster parameters as

$$E_{X_5} = E_{t_{2g}} - 3(dd\sigma) - (dd\delta), \quad (2)$$

$$E_{\Gamma'_{25}} = E_{t_{2g}} + 3(dd\sigma) + 4(dd\pi) + 5(dd\delta), \quad (3)$$

$$E_{X_3} = E_{t_{2g}} + 3(dd\sigma) - 4(dd\pi) - 3(dd\delta), \quad (4)$$

where $E_{t_{2g}}$ is the energy of the degenerate t_{2g} orbitals and X_5, Γ'_{25}, and X_3 are the irreducible representations of the space group that label the spatial symmetry of the single-particle states [14]. In general, one has, in the order of descending magnitude, $(dd\sigma) < 0$, $(dd\pi) > 0$, and $(dd\delta) < 0$. In the following calculations we use a set of single-particle parameters based on our previous work on some $3d$ transition-metal systems [11,12,15]: $(dd\sigma) = -1.0$ (its magnitude is chosen as the energy unit in the following calculations), $(dd\pi) = 0.7$, $(dd\delta) = -0.15$, and $E_{t_{2g}} = -1.0$. The corresponding single-particle bandwidth is $W = 8.5$. Two points are worth mentioning here. First, these are the renormalized parameters with only nearest-neighbor interactions among d orbitals explicitly included, not the "bare" band parameters as one would get in an all-electron single-particle tight-binding fit. Second, it should be pointed out that exact ratios of the single-particle parameters do not affect the qualitative physics studied in this work. In fact, we have tested several different sets of parameters; they all yield similar results.

The interaction term V^{dd} can be described by two parameters: the direct Coulomb integral U and the exchange integral J. The Coulomb energy for two particles on different d orbitals is $U - 2J$. We choose the ratio $U : J = 5$ (Ref. [9]). Again, the exact value of this ratio does not affect the qualitative physics studied in this work. This leaves U the only variable parameter in the present formulation. Below we will systematically study the effect of the d-d interaction U on the behavior of the spectral function evolution.

We consider the system with d^1 configuration, i.e., four electrons in the four-site tetrahedron cluster. Since we retain three orbitals per site per spin, there are 24 orbitals in the cluster (equivalent to a 12-site single-band Hubbard model). Simple combinatorial argument yields 10 626 many-body states in the neutral state of the cluster. After the application of the particle-number, spin, and space-group symmetry projections, the largest Hamiltonian matrix is of order 272, a significant reduction from the original ($10\,626 \times 10\,626$) matrix. All matrices are numerically diagonalized, and all eigenvalues and eigenstates are obtained and stored.

The spin-resolved photoemission spectral function is defined as

$$F_{\mathrm{PE}}(\omega, \sigma) = \sum_{\mu} \sum_{k} |\langle \phi_k^{N-1} | c_{\mu\sigma} | \phi_0^N \rangle|^2 \times \delta[\omega - (E_k^{N-1} - E_0^N)], \quad (5)$$

where ϕ_0^N and ϕ_k^{N-1} are the N-electron ground state and the kth $(N-1)$-electron final state, with energies E_0^N and E_k^{N-1}, respectively. The operator $c_{\mu\sigma}$ destroys an electron with spin σ and orbital index μ.

Figure 1 shows the calculated spin-resolved and integrated photoemission spectral functions for various values of U. Notice that in this work all energies are measured in units of $|(dd\sigma)|$. It is clearly seen that, as the value of U increases, the spectral function gradually evolves from a single-peak structure (the single-particle results) to a double-peak structure with a significant amount of spectral weight transferred to the "incoherent" part, i.e., beyond the range predicted by the single-particle theory. This is qualitatively different from the results of Monte Carlo simulation [16] and exact diagonalization [17] studies of a single-band Hubbard model. In those works it is found that with increasing interaction the upper and lower Hubbard bands are shifted away from the Fermi energy rather *rigidly* without any noticeable spectral weight transfer between coherent and incoherent parts. This demonstrates that the experimentally observed spectral weight transfer [8] is due to *combined* single-particle and many-body effects. A detailed analysis of the calculated results yields the following conclusions:

(i) When the interaction is weak, the spectral weight transfer is dominated by the *direct Coulomb interactions* since there is a large overlap between the majority-spin

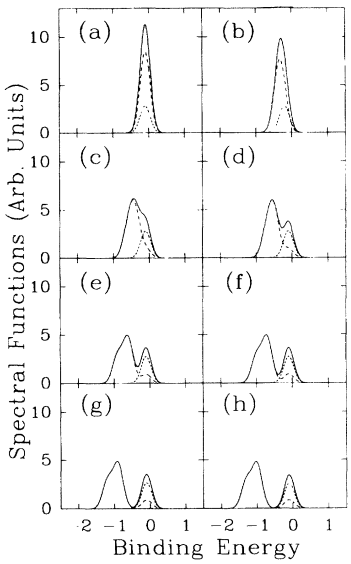

FIG. 1. The calculated spin-resolved photoemission spectral functions. Results in (a)–(h) correspond to interaction parameters (energies are measured in units of $|(dd\sigma)|$) U = 0.0, 1.0, 2.0, 3.0, 4.0, 5.0, 8.0, and 10.0. The solid, long-dashed, and short-dashed lines represent total, majority-spin and minority-spin results.

and minority-spin spectra [see Figs. 1(a)–1(d)]. The corresponding spectral function evolution is therefore very sensitive to the value of U, yielding drastic changes in the spectra for relatively small changes in U. For larger values of U, the direct overlap between the spectra of opposite spin orientations is small, and the main driving force behind the spectral weight transfer is the exchange interaction J. The peak positions in the spectra become less sensitive to changes in U. We compare the calculated results shown in Figs. 1(c)–1(e) to the "unusual" experimental data [8] for VO_2, $SrVO_3$, and $LaTiO_3$. These three systems all *have almost identical single-particle spectra,* which ensures a meaningful comparison with the calculated results. An excellent qualitative agreement is evident. A quantitative agreement is achieved with a detailed Hamiltonian parameter fit [18]. However, we want to emphasize the qualitative physics involved here, i.e., the observed "unusual" spectral function evolution is driven by the *d-d* interaction in a *realistic band environment.* In other words, it is due to *combined* single-particle and many-body effects.

(ii) The spectral weight in the coherent part, defined as the spectra with binding energy $E > -0.5$, and the incoherent part are calculated, and the results are shown in Figs. 2 and 3. These results agree qualitatively with the experimental measurements [8]. Again, a quantitative agreement is achieved with a detailed Hamiltonian parameter fit. One interesting observation is that the total spectral weight of *both* peaks decreases as U becomes large. The "missing" spectra actually are driven by strong direct Coulomb interaction to higher binding energies, which are not shown in Fig. 1.

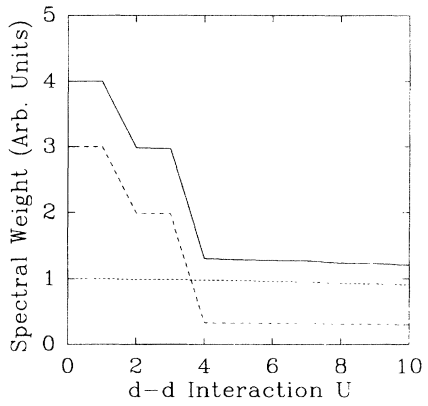

FIG. 2. The calculated spectral weight in the "coherent part," defined as $E > -0.5$. Energies are measured in units of $|(dd\sigma)|$. The solid, long-dashed, and short-dashed lines represent total, majority-spin, and minority-spin results.

(iii) The spectra in the incoherent part are fully polarized in majority-spin orientation, while the peak near the Fermi energy (the coherent part) is highly polarized in minority-spin orientation. The spin polarization is the result of electron correlation effects. It should be easily detectable in spin-resolved measurements. However, the degree of polarization may be modified slightly in real materials due to contributions from other electrons even if the *d*-electron contribution is well identified.

(iv) In Figs. 1(e)–1(h) it can be seen that the peak in the incoherent part has a shoulder on the high binding-energy side. This is due to the splitting of previously degenerate (or almost degenerate) levels. In other words, the incoherent part actually has a multipeak structure.

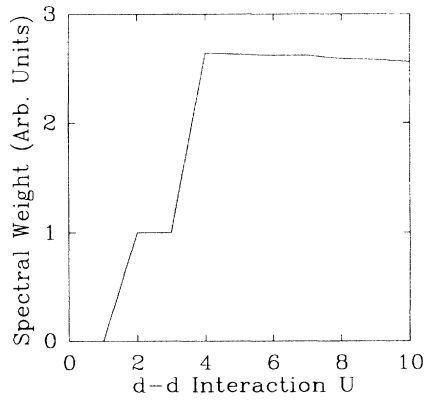

FIG. 3. The calculated spectral weight in the "incoherent part" with binding energies $-2.0 < E < 0.5$. Energies are measured in units of $|(dd\sigma)|$. The spectrum is fully polarized in the majority-spin orientation in this energy range.

In summary, we have studied the evolution of the photoemission spectral function of a realistic multiband d-electron model with d^1 configuration in an exact diagonalization approach. Results qualitatively different from those of single-particle and single-band Hubbard models have been obtained. They provide a natural explanation for the "unusual" spectral weight transfer recently observed in several d-electron materials with various ratios of interaction to single-particle bandwidth. The conclusions on the combined single-particle and many-body effects in strongly correlated systems drawn from the calculated results are expected to be valid for other similar systems with degenerate occupied orbitals.

This work was supported in part by the National Science Foundation and a supercomputer grant from the National Supercomputing Center for Energy and the Environment at University of Nevada, Las Vegas, Nevada.

[1] See, for example, E. Dagotto, Rev. Mod. Phys. **66**, 763 (1994), and references therein.

[2] A. K. McMahan, R. M. Martin, and S. Satpathy, Phys. Rev. B **38**, 6650 (1988).

[3] M. S. Hybertsen, M. Schlüter, and N. E. Christensen, Phys. Rev. B **39**, 9028 (1989).

[4] L. F. Mattheiss and D. R. Hamann, Phys. Rev. B **40**, 2217 (1989).

[5] M. J. De Weert, D. A. Papaconstantopoulos, and W. E. Pickett, Phys. Rev. B **39**, 4235 (1989).

[6] M. S. Hybertsen, E. B. Stechel, M. Schlüter, and D. R. Jennison, Phys. Rev. B **41**, 11 068 (1990).

[7] M. M. Steiner, R. C. Albers, and L. J. Sham, Phys. Rev. B **45**, 13 272 (1992); W. Nolting *et al., ibid.* **40**, 5015 (1989).

[8] A. Fujimori, I. Hase, H. Namatame, Y. Fujishima, Y. Tokura, H. Eisaki, S. Uchida, K. Takegahara, and F. M. F. de Groot, Phys. Rev. Lett. **69**, 1796 (1992).

[9] C. Herring, in *Magnetism,* edited by G. T. Rado and H. Suhl (Academic, New York, 1966), Vol. 4.

[10] J. C. Slater and G. F. Koster, Phys. Rev. **94**, 1498 (1954).

[11] Changfeng Chen, Int. J. Mod. Phys. B **5**, 1147 (1991), and references therein.

[12] Changfeng Chen, Phys. Rev. B **45**, 13 811 (1992); **48**, 1318 (1993); J. Phys. Cond. Matter **4**, 9855 (1992).

[13] The choice of a bulk fcc lattice has no particular significance here. The qualitative behavior of the calculated interaction-driven spectral function evolution should not depend on the lattice structure [see, for example, H. Eskes, M. B. J. Meinders, and G. A. Sawatzky, Phys. Rev. Lett. **67**, 1035 (1991); J. Callaway, J. W. Kim, L. Tan, and H. Q. Lin, Phys. Rev. B **48**, 11 545 (1993)].

[14] For the group-theoretical analysis of the cluster model of fcc structure, see A. Reich and L. M. Falicov, Phys. Rev. B **37**, 5560 (1988). Note, however, an eight-site cluster model is employed in that work. We retain only the Γ and X representations for the present four-site cluster model.

[15] Changfeng Chen, Phys. Rev. B **41**, 1320 (1990).

[16] S. R. White, D. J. Scalapino, R. L. Sugar, and N. E. Bickers, Phys. Rev. Lett. **63**, 1523 (1989).

[17] E. Dagotto, R. Joynt, A. Moreo, S. Bacci, and E. Gagliano, Phys. Rev. B **41**, 9049 (1990); Q. Li, J. Callaway, and L. Tan, Phys. Rev. B **44**, 10 256 (1991).

[18] Because of differences in their band structures, slightly different single-particle parameters are required to fit the spectra for the three systems used in the experiment. Since it is not clear to what degree the contribution from other electrons is mixed in the spectra nominally assigned as "d-electron spectra," we do not want to emphasize the detailed numerical assignment (i.e., the *effective* values) of the d-electron parameters, which will be reported elsewhere. Rather, we stress the qualitative physics that is expected to be valid in general in all similar systems.