Elementary excitations in the coupled spin-orbital model

J. van den Brink,* W. Stekelenburg, D. I. Khomskii, and G. A. Sawatzky
Laboratory of Applied and Solid State Physics, Materials Science Centre, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands

K. I. Kugel
Scientific Center for Applied Problems of Electrodynamics, Russian Academy of Science, Izhorskaya Street 13/19, Moscow 127412, Russia

(Received 10 February 1998; revised manuscript received 7 May 1998)

The elementary excitations of a model Hamiltonian that captures the low-energy behavior of a simple twofold-degenerate Hubbard Hamiltonian, with Hund's rule coupling, is studied. The phase diagram in the mean-field limit and in a two-site approach reveals a rich variety of phases where both the orbital and the spin degrees of freedom are ordered. We show that, besides the usual spin waves (magnons), there also exist orbital waves (orbitons) and, most interestingly, in a completely ferromagnetically coupled system, a combined spin-orbital excitation which can be visualized as a bound state of magnons and orbitons. For a completely degenerate system the bound states are found to be the lowest-lying elementary excitations, both in one and two dimensions. Finally we extend our treatment to almost-degenerate systems. This can serve as an example that elementary excitations in orbitally degenerate strongly correlated electron systems in general carry both spin and orbital character. [S0163-1829(98)10839-1]

I. INTRODUCTION

The d and f wave functions of free atoms or ions are, besides Kramers (spin) degenerate, also fivefold and sevenfold orbital degenerate. In crystals this degeneracy may be lifted by the crystal field (interactions with the ligands). There are, however, interesting situations where this degeneracy is only partially lifted, so that in a high-symmetry situation d or f levels remain degenerate. The best known examples are Cu^{2+} (d^9), Mn^{3+} (d^4), and Cr^{2+} (d^4) in cubic symmetry (in an octahedral surrounding). According to the famous Jahn-Teller theorem,[1] this degeneracy should be lifted in the ground state. In a concentrated system this leads to a phenomenon known as the cooperative Jahn-Teller effect, or orbital ordering.[2,3] An interesting aspect of this phenomenon is the strong interplay between the orbital and spin (magnetic) ordering. The orbital occupation determines the character of the magnetic exchange interaction (the Goodenough-Kanamori-Anderson rules),[4] and, vice versa, modification of the magnetic structure, e.g., by an external field, may change orbital occupation and lead to a change in the crystal structure.[3,5]

The existence of orbital degrees of freedom, strongly interacting with the spins, not only determines the orbital and spin structure in the ground state. It should also have important consequences for the elementary excitations of such systems. Thus, in addition to the collective excitations of the magnetic subsystem—magnons or spin waves—orbital excitations—orbitons—also may exist in this case. This was pointed out in Ref. 3, and was studied for a specific model for the manganites in Ref. 6. Because of the intimate connection of spin and orbital degrees of freedom, one can also expect strong interaction and possible mixing of these two types of excitations. This problem was not addressed until now, and it is one of the aims of the present study.

The systems with orbital degeneracy form a rather special class of compounds with very interesting and rich properties. Among them are, in particular, many compounds containing Cu^{2+} (such as high-T_c superconductors), manganites of the type $La_{1-x}Sr_xMnO_3$ with a colossal magnetoresistance, and many other transition-metal compounds containing vanadium ($LiVO_2$, CaV_4O_9),[7,8] trivalent nickel ($PrNiO_3$),[9] etc. The study of the elementary excitations in these compounds, besides being of interest in itself, may shed some light on their unusual properties.

In this work we consider the typical situation of materials containing localized electrons with spin $S=\frac{1}{2}$ with doubly degenerate orbitals. We can describe this situation by the doubly degenerate Hubbard model

$$H=H_t+H_U+H_J, \tag{1}$$

with

$$H_t=\sum_{\langle ij\rangle,\sigma,\alpha,\beta} t_{ij}^{\alpha\beta} c_{i\alpha\sigma}^{\dagger} c_{j\beta\sigma}, \tag{2}$$

$$H_U=\sum_{i,\sigma,\sigma',\alpha,\beta} U^{\alpha\beta} n_{i\alpha\sigma} n_{i\beta\sigma'} (1-\delta_{\alpha,\beta}\delta_{\sigma,\sigma'}), \tag{3}$$

$$H_J=\sum_{i,\alpha,\beta} -J_H^{\alpha\beta} \mathbf{S}_{i\alpha}\cdot\mathbf{S}_{i\beta}(1-\delta_{\alpha,\beta}), \tag{4}$$

in which, besides the usual terms (electron hopping and on-site Coulomb repulsion), we also added the on-site (Hund) exchange interaction.

In realistic situations the hopping matrix elements $t_{ij}^{\alpha\beta}$ depend on the type of orbital involved.[3–6] This leads to enormous technical complications. As here we are interested in

the basic typical features of the excitation spectrum of our system, we treat a simplified symmetric model, assuming

$$t_{ij}^{\alpha\beta}=t_{ij}\delta_{\alpha\beta}, \tag{5}$$

keeping only the nearest-neighbor hopping for equal orbitals. At the end of this paper we comment upon which modifications of our results should occur for a more general choice of hopping integrals. In the case of strong interaction $t \ll U$, only the degrees of freedom of the localized electrons with spin $\frac{1}{2}$ and the orbitals are left. Consequently we can reduce the electronic model [Eq. (1)] to a model describing coupled spins and orbitals. For the doubly degenerate case we can describe the orbital degrees of freedom by an effective pseudospin $T=\frac{1}{2}$, so that, e.g., an occupied orbital α corresponds to $T^z=\frac{1}{2}$ and orbital β to $T^z=-\frac{1}{2}$. The effective Hamiltonian has the generic form

$$H=-J_s\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j-J_t\sum_{\langle ij\rangle}\mathbf{T}_i\cdot\mathbf{T}_j-4J_{st}\sum_{\langle ij\rangle}(\mathbf{S}_i\cdot\mathbf{S}_j)\ (\mathbf{T}_i\cdot\mathbf{T}_j), \tag{6}$$

where for the model with $t_{ij}^{\alpha\beta}$ given by Eq. (5) the exchange constants have definite values.[3] Thus for this particular choice of hopping matrix elements and for vanishing Hund's rule exchange the model (1) reduces to a double-spin Heisenberg model.

The characteristic feature of our situation is the existence and strong interplay of ''spins'' S and T which emerge due to the third term of Hamiltonian (1). As we shall see, this interaction leads to a significant mixing of spin and orbital degrees of freedom, giving rise, even, to the possibility of the formation of spin-orbiton bound states. Note that the form of the interaction between the spins and orbitals in Eq. (1) is a consequence of the Coulomb-Hund's rule interaction between electrons, and is different from what one expects from simply coupling two Heisenberg spin systems. As we want to study the generic features of the coupled spin-orbital system, we consider a general case, treating arbitrary values of the exchange parameters J_s, J_t, and J_{st}. It is explicitly shown that a spin-orbital bound state exists when all exchange parameters are ferromagnetic, and we show that the same mechanism that leads to a bound state for ferromagnetic exchange parameters is also operative in a system with antiferromagnetic exchange parameters.

Notice that in the general situation with Hund's rule exchange included and with realistic values of the hopping integrals $t_{ij}^{\alpha\beta}$, the resulting spin-orbital Hamiltonian of type (6) can contain terms anisotropic in the T operators,[3] and even terms linear in T. However, even with the more realistic hopping integrals there may exist situations[10] in which the symmetry in orbital space remains similar to that of Hamiltonian (6). For simplicity we consider such a symmetric double-Heisenberg model because, as it turns out, the conditions for the existence of the bound spin-orbital excitations are the most stringent exactly in this case, and if, as we will show, they exist in this case, it will be even more so with the T anisotropy taken into account.

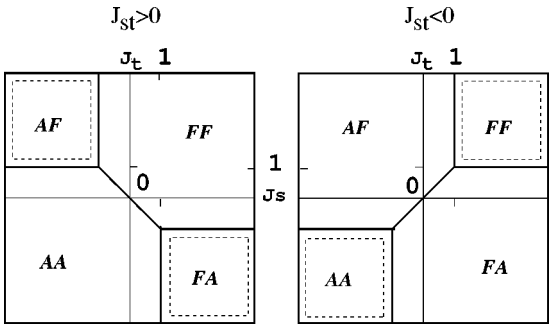

FIG. 1. Phase diagram of the model Hamiltonian in the mean-field approximation. FF: both spin and pseudospin are ferromagnetically ordered; FA: spin ferro, pseudospin antiferro; AF: spin antiferro, pseudospin ferro; AA: both spin and pseudospin are antiferro ordered. J_s and J_t are in units of $|J_{st}|$.

II. GROUND STATE

If our aim is to calculate the elementary excitations of a system described by the Hamiltonian (6), we first have to know the ground-state wave functions. We can easily calculate a phase diagram within two different approximations: the mean-field and the two-site approximation. In the latter only two interacting spins and pseudospins are considered, so that the ground state is described in a way similar to a valence-bond state, which is a good starting point for low-dimensional spin systems. Starting from ground-state wave functions obtained within one of these approximations, one can test the stability of the ground state by calculating its response to, for instance, spin waves and combined orbital and spin excitations.

A. Mean-field solution

A common method used to gain some understanding of a Hamiltonian is a mean-field approximation. In our case it is possible to separate spin and orbital degrees of freedom by replacing operators we want to exclude from our consideration by the averages of their correlation function that appears in the Hamiltonian. In this way we generate two mean-field Hamiltonians

$$H_s^{MF}=-J_s\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j-4J_{st}\sum_{\langle ij\rangle}\langle\mathbf{T}_i\cdot\mathbf{T}_j\rangle\mathbf{S}_i\cdot\mathbf{S}_j=-J_s'\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j \tag{7}$$

and

$$H_t^{MF}=-J_t\sum_{\langle ij\rangle}\mathbf{T}_i\cdot\mathbf{T}_j-4J_{st}\sum_{\langle ij\rangle}\langle\mathbf{S}_i\cdot\mathbf{S}_j\rangle\mathbf{T}_i\cdot\mathbf{T}_j$$

$$=-J_t'\sum_{\langle ij\rangle}\mathbf{T}_i\cdot\mathbf{T}_j. \tag{8}$$

In Hamiltonian (7) the orbital degrees of freedom are integrated out, and in Hamiltonian (8) the average over the spin degrees of freedom is taken. In this way the spin and orbital degrees of freedom are decoupled. After doing the mean-field averaging[6] we have, in some sense, thrown away the interesting part of our problem and returned to a renormalized Heisenberg model, where the ground-state properties and elementary excitations are well known. Neverthe-

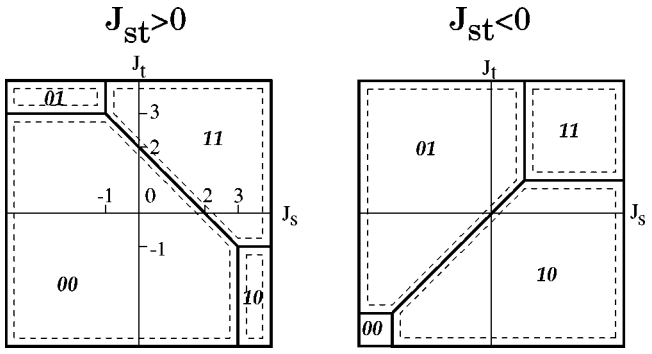

FIG. 2. Phase diagram of the two-site solution of the model Hamiltonian. In phase 11, both spin and pseudospin are in a triplet state; in phase 10, the spin is triplet and pseudospin singlet; in 01 phase the spin is singlet, and the pseudospin is triplet; and in 00 phase both the spin and pseudospin are in a singlet state. J_s and J_t are in units of $|J_{st}|$.

less, as in a first approximation this is still a useful approach. The phase diagram in this approximation is given in Fig. 1.

B. Two-site solution

In a mean-field approximation the short-range interactions are averaged out, and if we want to go beyond this approximation and gain insight into local properties it is more useful to consider a few interacting particles. This is especially important here because the transformation properties of these terms of Hamiltonian (6) are different, and a more accurate account of the real singlet correlations (as compared to the mean field, essentially Ising-like, treatment) is essential. To this end case we consider two spins and two pseudospins, and obtain the Hamiltonian

$$H_{1,2} = -J_s \mathbf{S}_1 \cdot \mathbf{S}_2 - J_t \mathbf{T}_1 \cdot \mathbf{T}_2 - 4J_{st}(\mathbf{S}_1 \cdot \mathbf{S}_2)(\mathbf{T}_1 \cdot \mathbf{T}_2). \quad (9)$$

This simple case can be treated exactly, and using these results, we obtain the modified phase diagram. The combinations of (pseudo)spin operators can either be singlet or triplet, and the ground-state energy of the various combinations correspond to the different phases in Fig. 2, for $J_{st}>0$ and $J_{st}<0$, respectively. The character of the first excited state for the various phases for $J_{st}>0$ is shown in Fig. 3. A similar picture can be made for $J_{st}<0$. In spite of the relative simplicity of the Hamiltonian, there is a rich variety of ground and first excited states, even in the two-site approach.

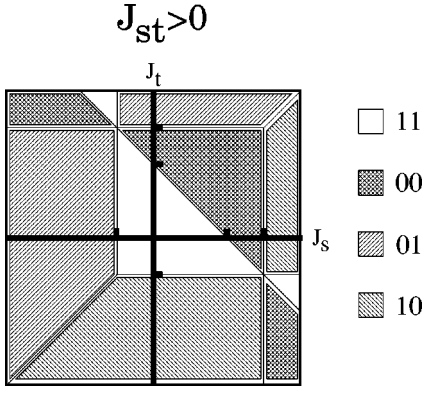

FIG. 3. Character of the first excited state in the two-site system. J_s and J_t are in units of $|J_{st}|$.

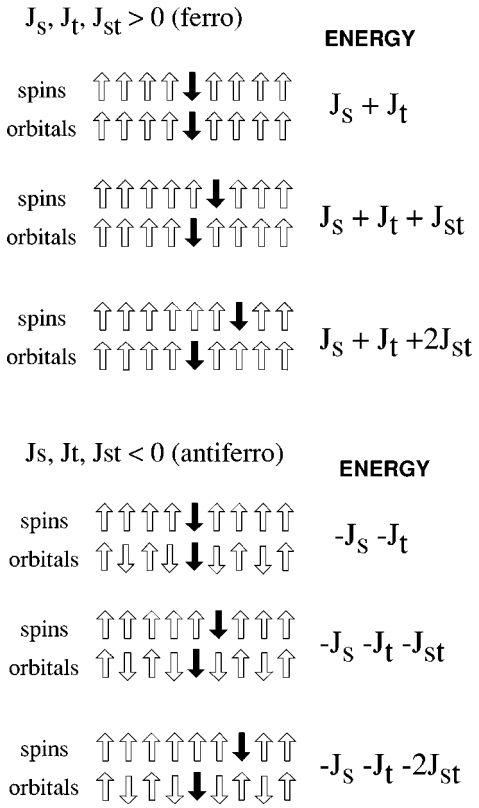

FIG. 4. Excitation energy for Ising spins and pseudospins for a completely ferro and a completely antiferro 1D system in the regime where $J_{st}<J_t<J_s$. When the spin and orbital are close together, the excitation energy is lower than in the case when they are far apart.

In the white, central part of this figure, the ground state is singlet, both in the spins and orbitals. The first excited state, however, is not just the change of one singlet into a triplet, but corresponds to the state where both the spins and orbitals are in a triplet configuration. This is due to the interaction between the spin and orbital degrees of freedom, and this very simple example shows that some of the excitations of a system with such an interaction, in this case the lowest one, carry both spin and orbital character.

III. FERRO-FERRO SYSTEM

When J_s, J_t, and J_{st} are chosen in a range where both the spins and pseudospins order ferromagnetically, a ferro-ferro phase, we can obtain exact analytical expressions for the elementary excitations. In a ferromagnetic Heisenberg model the exact ground state is the state with all spins pointing in the same direction, as opposed to a Heisenberg Hamiltonian with an antiferromagnetic exchange where quantum fluctuations affect the Néel spin order. The coupling between the spins and pseudospins can lead to the formation of bound states of spin and pseudospin excitations. This is illustrated for a one-dimensional (1D) ferro-ferro system in the upper part of Fig. 4. Let us consider Ising spins (and pseudospins), and examine the energy of an excitation of one spin plus one pseudospin. If the two excited spins are far away from each

other, the part of the excitation energy due to the spin pseudospin interaction [the third term in Eq. (6)] is $2J_{st}$. When the excited spins are close to each other, on the nearest-neighbor sites or on the same site, this excitation energy is J_{st} and 0, respectively. The same argument holds if the system has antiferromagnetic exchanges. In the lower part of Fig. 4 the same attraction between the spin and the orbital excitation is found for a completely antiferromagnetically coupled system in the regime where $|J_{st}|>|J_t|>|J_s|$ (see the phase diagrams Figs. 1 and 2). One can easily check that the situation is the same also for other types of ordering. This means that a magnon and an orbiton (orbital wave) have an attractive interaction and that, in principle, depending on the dimensionality and strength of the attraction, the magnon and orbiton can form a bound state.

A. Equations of motion

In order to establish whether it is indeed possible to obtain bound states that are combinations of orbital and spin waves in the excitation spectrum of the model system, we use the equation of motion method, which provides a simple, and in the ferromagnetic case exact, way to calculate bound-state energies. Before addressing this issue, let us first examine a single (pseudo)spin excitation.

Starting from Hamiltonian (6) and with a ground state $|0\rangle$ with energy E_0 where all (pseudo)spins are aligned in the positive-z direction, a single spin is excited (the derivation for single orbital excitations is equivalent). The equation of motion for this excitation is

$$(H-E_0)S_m^-\ |0\rangle \equiv \omega_s S_m^-|0\rangle=[H,S_m^-]|0\rangle. \tag{10}$$

The evaluation of the commutator and transformation to Fourier-space yields the following Goldstone modes:

$$\omega_s(\mathbf{Q})=2(J_s+J_{st})\sum_{\mathbf{a}}\ (1-\cos\mathbf{Q}\cdot\mathbf{a}), \tag{11}$$

$$\omega_t(\mathbf{Q})=2(J_t+J_{st})\sum_{\mathbf{a}}\ (1-\cos\mathbf{Q}\cdot\mathbf{a}), \tag{12}$$

where the lattice vectors are denoted by **a**. The presence of the coupling between the spin and orbital degrees of freedom, parametrized by J_{st}, thus merely renormalizes the spin/orbital wave spectrum.

The equation of motion for a combined spin and pseudospin excitation $S_m^-T_n^-$ is

$$(H-E_o)S_m^-T_n^-|0\rangle \equiv \omega S_m^-T_n^-|0\rangle=[H,S_m^-T_n^-]|0\rangle. \tag{13}$$

We find, for the Fourier transform of the equation of motion,

$$\{\omega-\gamma(\mathbf{Q},\mathbf{q})\}A(\mathbf{Q},\mathbf{q})$$

$$=-8J_{st}\left(\frac{a}{2\pi}\right)^d\sum_{\mathbf{a}}\ (\cos\mathbf{Q}\cdot\mathbf{a}/2-\cos\mathbf{q}\cdot\mathbf{a})$$

$$\times\int d\mathbf{k}(\cos\mathbf{Q}\cdot\mathbf{a}/2-\cos\mathbf{k}\cdot\mathbf{a})A(\mathbf{Q},\mathbf{k}), \tag{14}$$

with

$$A(\mathbf{Q},\mathbf{q})=S^-_{\mathbf{Q}/2-\mathbf{q}}T^-_{\mathbf{Q}/2+\mathbf{q}}|0\rangle, \tag{15}$$

$$\gamma(\mathbf{Q},\mathbf{q})=\omega_s(\mathbf{Q}/2+\mathbf{q})+\omega_t(\mathbf{Q}/2-\mathbf{q}), \tag{16}$$

where d is the dimensionality of the system. The total momentum of the excitation is **Q**, and the relative momentum of the combined spin and pseudospin excitation is **q**. In order to check if there is a self-consistent solution of the equation of motion, Eq. (14) is iterated once. Then a set of equations is found that can be represented in a matrix $|M_{\alpha,\beta}|$ of order d for hypercubic lattices. In a three-dimensional system $\alpha,\beta=x,y,z$. The set of equations has a solution if, and only if,

$$\mathrm{Det}|\,\delta_{\alpha,\beta}-M_{\alpha,\beta}|=0, \tag{17}$$

with

$$M_{\alpha,\beta}(\mathbf{Q})=-8J_{st}\left(\frac{a}{2\pi}\right)^d \times\int d\mathbf{k}\frac{(\cos Q_\alpha/2-\cos k_\alpha)(\cos Q_\beta/2-\cos k_\beta)}{\omega-\gamma(\mathbf{Q},\mathbf{k})}. \tag{18}$$

If the system is one dimensional the condition above reduces to

$$1=-\frac{8J_{st}}{2\pi}\int_{-\pi}^{\pi}dk\ \frac{(\cos Q/2-\cos k)^2}{\omega-\omega_s(k+Q/2)-\omega_t(k-Q/2)}, \tag{19}$$

where the lattice spacing is set to unity. This expression resembles the condition for the existence of a two-magnon bound state in a pure ferromagnetic Heisenberg model.[11]

B. Bound states

We restrict ourselves to the ferro-ferro phase, i.e., according to Fig. 2, $2J_{st}<J_s+J_t$ and $J_{st}>0$, and determine the bound-state energies from conditions (17) and (18). The integrals appearing in the latter equation are, as in the two-magnon problem, by no means trivial, and can only be determined analytically in special cases. In general these equations can be solved numerically.

Before examining the integrals in detail, let us first turn to the excitations that lie in the continuum. The continuum of excitations starts at the energy $\omega_c(\mathbf{Q})$, where the denominator of the matrix elements $M_{\alpha,\beta}$ starts to diverge. The condition for this is

$$\omega_c(\mathbf{Q})=\mathrm{Min}_{\mathbf{k}}[\,\epsilon_s(\mathbf{Q}/2+\mathbf{k})+\epsilon_t(\mathbf{Q}/2-\mathbf{k})]. \tag{20}$$

The single (pseudo)spin-wave excitation energy *always* lies in the two-particle continuum,

$$\omega_s(\mathbf{Q})\geqslant\omega_c(\mathbf{Q}),$$

$$\omega_t(\mathbf{Q})\geqslant\omega_c(\mathbf{Q}).$$

The explanation for this is that in a combined spin and orbital excitation the momentum is distributed over the two subsystems, and, because the excitation spectrum in the ferro-ferro case is nonlinear and concave ([1−cos]-like), the

energy of two excitations with smaller momenta can be lower than the energy of one excitation with large momentum.

First we consider the *one-dimensional* case. The integral in Eq. (19) can be reduced to the form

$$\int \frac{(a-\cos k)^2}{1+b\cos k+c\sin k}dk, \tag{21}$$

which is known analytically. The condition that the integral be equal to unity is given by the roots of a third-order polynomial. Not so much is gained following this procedure, as the resulting expressions are very long and complicated. So let us consider some special momenta in the Brillouin zone.

$\mathbf{Q}=\mathbf{0}$. For $Q=0$, condition (19) for a bound state at $\omega=0$ reduces to $2J_{st}=J_s+J_t$, which is exactly at the phase boundary of the ferro-ferro phase in the two-site phase diagram (Fig. 2). A bound state at negative ω appears when $2J_{st}>J_s+J_t$. This simply means that the ground state is not stable (by exciting it, energy is gained), which can be expected as the two-site phase diagram shows that the antiferro-antiferro phase is the ground state in this parameter range. The ferro-ferro ground state is found to be exactly stable in the parameter ranges shown in Fig. 2, indicating that the two-site approximation gives a good prediction for the ground-state spin and orbital order, whereas the mean-field solution fails to predict the right ordering.

$\mathbf{Q}=\pi$. By treating this special case, we can prove that there is *always* a bound state in the ferro-ferro system, at least in one dimension. When $Q_\alpha=\pi$ for all α, at the corner of the Brillouin zone, the equations simplify considerably. The off-diagonal matrix elements in Eq. (17) vanish, so that $M_{\alpha,\beta}=D\delta_{\alpha,\beta}$. This yields

$$\omega_b^{1d}(\pi)=2(J_s+J_t)-(J_s-J_t)^2/4J_{st}, \tag{22}$$

$$\omega_c^{1d}(\pi)=4(\mathrm{Min}[J_s,J_t]+J_{st}). \tag{23}$$

From these equations follows that $\omega_b^{1d}(\pi)<\omega_c^{1d}(\pi)$ for any J_s, J_t, and J_{st}, except when $J_{st}=|J_s-J_t|$, where $\omega_b^{1d}(\pi)=\omega_c^{1d}(\pi)$. This proves that at $Q=\pi$ there is always a bound state.

At $\omega(\mathbf{Q})\rightarrow\omega_c(\mathbf{Q})$ *in one dimension*. For energies approaching the continuum $[\omega(\mathbf{Q})\rightarrow\omega_c(\mathbf{Q})]$, the integrand in Eq. (19), diverges as k^{-1} (except for $Q=0$), making the integral logarithmically divergent. We can conclude from this that for any Q (except for $Q=0$) there is always a point between $-\infty<\omega<\omega_c(Q)$ where the integral is equal to unity.

From the considerations above we can conclude that for the one-dimensional system in the range $0<Q\leqslant\pi$ a spin-orbital bound state always exists, and that this bound state is the lowest-lying elementary excitation of the ferro-ferro phase of model (6). It is by definition lower in energy than the spin-orbital continuum, which in turn is lower than the single (pseudo) spin excitations. Before illustrating the statement above with numerical examples, let us consider one more special case, namely:

$\mathbf{J_s}=\mathbf{J_t}=\mathbf{J_{st}}$. For these parameters the system is exactly at the phase boundary of the ferro-ferro ground state. Now Eq. (19) takes a particularly simple form, and the solution for the bound state for the one-dimensional system is

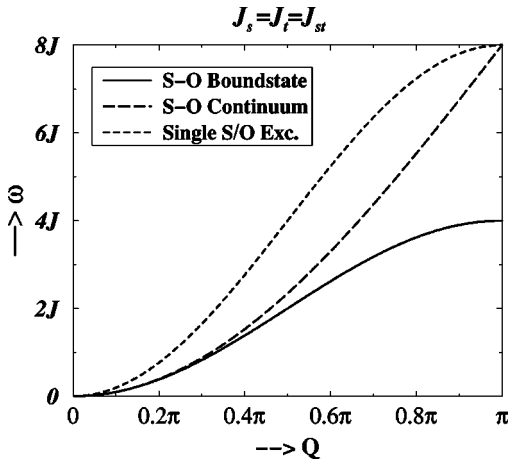

FIG. 5. Dispersion of the spin-orbital bound state, spin-orbital continuum, and single-spin and single-orbital excitations in a one-dimensional ferro-ferro system where $J_s=J_t=J_{st}$. The single spin and single orbital excitation energies coincide in this case. The unit of energy is $J=J_s$.

$$\omega_b^*(Q)=2J_{st}(1-\cos Q). \tag{24}$$

For the lower bound of the continuum, the single-spin and single-orbital excitations, in this case one obtains

$$\omega_c^*(Q)=8J_{st}(1-\cos Q/2), \tag{25}$$

$$\omega_s^*(Q)=\omega_t^*(Q)=4J_{st}(1-\cos Q). \tag{26}$$

In Fig. 5 the dispersion relations of these excitations are shown. The spin-orbital bound state is always the lowest energy excitation of the system. Note that for small Q the spin-orbital continuum and the bound state are very close in energy, and their energy difference is of the order of $(J_{st}/16)Q^4$.

The numerical solution of the bound-state equation for a one-dimensional system is shown in Fig. 6 for two parameter sets. For small momenta the bound state is always very close to the two-particle continuum. When J_{st} is reduced, the single spin, single orbital, and continuum shift down in energy, approaching the bound-state energy. This can be expected, since in the case when $J_{st}=0$ and $J_s=J_t$ the lower bound of the continuum and the single-spin and single-orbital excitation spectra all coincide.

In the right part of Fig. 6 a case is shown where the single-spin and single-orbital excitations have a different dispersion, i.e., $J_s\neq J_t$. The single-orbital excitation is shifted up in energy with respect to the single-spin excitation. The bound state, continuum, and magnon excitations are close in energy, and in the limit that $J_t\rightarrow\infty$ all three coincide, as can be expected.

In Fig. 7 a typical result for a two-dimensional system is shown. It is found numerically that there always exists at least one bound state, also in two dimensions. Similar to the 1D case, the bound state is only well separated from the continuum at the edge of the Brillouin zone. If $J_s=J_t$ it can be shown that

$$\omega_b(\pi,0)=4(2J_s+2J_{st}-\sqrt{2J_{st}^2+2J_sJ_{st}+J_s^2}) \tag{27}$$

and

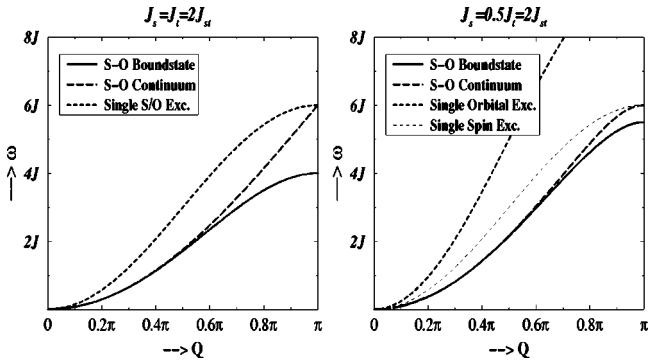

FIG. 6. Dispersion of the spin-orbital bound-state, spin-orbital continuum, and single-spin and single-orbital excitations in a one-dimensional ferro-ferro system. The unit of energy is $J=J_s$.

$$\omega_b(\pi,\pi)=4(2J_s+J_{st}), \tag{28}$$

so that at these points the bound state is considerably lower than the continuum.

C. Almost degeneracy

In the subsections above we assumed that the two atomic levels are completely degenerate. Crystal fields generally split the two levels. Within the approach above it is fairly easy to treat this energy splitting. Suppose the energy difference between the orbitals α and β is Δ. The Hamiltonian to be added to Eq. (1) is

$$H_\Delta=\Delta/2\sum_{i,\sigma}(n_{i,\sigma,\beta}-n_{i,\sigma,\alpha})=\Delta\sum_i T_i^z, \tag{29}$$

where the last equality follows from the definition of the T operators.

The level splitting manifests itself in the pseudospin language as a magnetic field for the orbitals. Carrying through the calculation for the excitations leads to renormalized spin waves, orbital waves, and bound states:

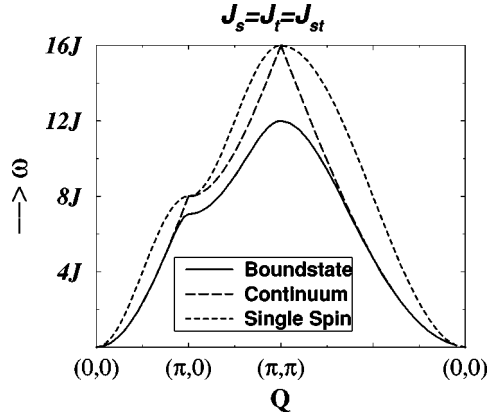

FIG. 7. Dispersion of the spin-orbital bound state and spin-orbital continuum, and single-spin and single-orbital excitations in a two-dimensional ferro-ferro system. The unit of energy is $J=J_s$.

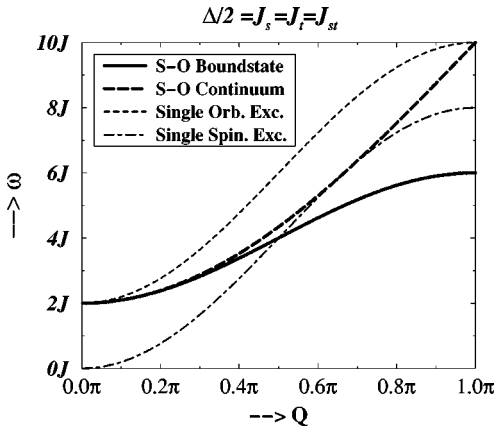

FIG. 8. Dispersion of the spin-orbital bound state, spin-orbital continuum, and single-spin and single-orbital excitations for an almost degenerate 1D ferro-ferro system where $\Delta/2=J_s=J_t=J_{st}$. The unit of energy is $J=J_s$.

$$\overline{\omega}_s=\omega_s,$$

$$\overline{\omega}_t=\omega_t+\Delta,$$

$$\overline{\omega}_{st}=\omega_{st}+\Delta. \tag{30}$$

The spin-wave spectrum is not affected, but the ''magnetic field'' for the orbitals causes a gap in the orbital excitation spectrum. This can be expected since, due to the magnetic field, there is no breaking of a continuous symmetry in the orbital case, and hence no Goldstone mode. This is reflected in the bound-state energy being gapped. The equalities above permit a convenient generalization of the results derived for the fully degenerate system to the situation where the levels are nondegenerate or almost degenerate, as illustrated in Fig. 8, where the dispersion of the bound state in the case of an orbital energy splitting of $\Delta=2J_s$ is shown.

From the results in this section one can also understand what will be the situation in the general case discussed at the end of Sec. I. That is, the orbital part of the effective-spin pseudospin Hamiltonian is in the general case anisotropic, containing both terms of the type (29) and Ising-like terms $T_i^zT_j^z$. The situation then will be simular to the one discussed above: there will appear a gap in the orbiton spectrum (see also Ref. 6). The spin-orbital bound state can still exist below the combined spin-orbital continuum (but in general above pure spin waves), and one can show that the conditions for their existence will be even less stringent than in the case of gapless orbitons. However, as can be seen from Fig. 8, these bound states will not be the lowest excitations in the system, at least not in the whole Brillouin zone. Nevertheless they will lie lower than the orbital waves themselves, and they definitely have to be taken into account in a general treatment of properties of such systems. Note also that the orbital spectrum may still be gapless, even with realistic hopping integrals $t_{ij}^{\alpha\beta}$ due to the orbital-lattice symmetry; this can lead to particular features of such systems like quantum disorder;[10] the nature of the elementary excitations plays a crucial role in this problem.[12]

IV. DISCUSSION

We studied the low-energy excitations of a twofold-degenerate Hubbard model in the strong-coupling limit with, on average, one electron per site and symmetric hopping integrals. We observe that, besides the separate spin and orbital excitations, combined excitations (spin-orbiton bound states) can exist, and can even be the lowest-lying elementary excitation of the system. In general overlap integrals depend explicitly on the symmetry of the orbitals; this, together with Hund's rule coupling, may break the continuous rotational symmetry of the orbital channel. In this case one can expect that the eventual spin-orbital bound states, which are gapless in the simplest case, become gapped and may be the lowest-lying excitation only at larger momenta.

If not all interactions are ferromagnetic, there is *a priori* no reason to expect that the orbital and spin-orbital bound states behave qualitatively different from the ferro case, although the quantum fluctuations might, or might not, destroy long-range order.[10,12,13] This interesting aspect of the system with antiferromagnetic interactions still deserves further study.

The low-energy collective modes of the orbitally degenerate Hubbard model certainly contribute to the thermodynamic properties of the system, and should be observable in, for instance, susceptibility and specific-heat measurements. It should be stressed, however, that elementary excitations with predominantly orbital character are in general gapped, and therefore lead only to moderate changes in thermodynamic quantities. Experiments that are sensitive to higher-energy scales might give direct evidence for the existence of orbitons and spin-orbital bound states. Orbital excitations, however, are strongly coupled to phonons, and it might be difficult to distinguish between these two contributions in experiment. One of the possibilities to pin them down may be connected with the possible anomalies of the phonon-dispersion relations induced by the orbital degrees of freedom. Another possibility might be that, since the orbital excitations also couple to the spin degrees of freedom, variations in the spin order, for instance induced by an external magnetic field, will be reflected in the energy and dispersion of the orbital related excitations, and consequently in the properties of the phonons mixed with orbitons.

In conclusion, we studied a model Hamiltonian that captures the low-energy behavior of a twofold-degenerate Hubbard Hamiltonian. We presented the phase diagram in the mean-field limit and in a two-site approach, revealing a rich variety of phases where both the orbital and spin degrees of freedom can be ordered (anti)ferromagnetically. We have shown that in this case there may exist, besides the usual spin waves (magnons), orbital waves (orbitons) and, most interestingly, the combined spin-orbital excitation which can be visualized as bound states of magnons and orbitons. In a fully degenerate system the bound states are found to be the lowest-lying elementary excitations, both in one and two dimensions. This shows that the elementary excitations in orbitally degenerate strongly correlated electron systems in general may carry both spin and orbital character.

ACKNOWLEDGMENTS

This work was financially supported by the Nederlandse Stichting voor Fundamenteel Onderzoek der Materie (FOM) and the Stichting Scheikundig Onderzoek Nederland (SON), both financially supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO). J.vdB. acknowledges with appreciation the support by the Alexander von Humboldt-Stiftung, Germany.

*Present address: Max Planck Institut FKF, Heisenbergstr. 1, 70569 Stuttgart, Germany. Electronic mail: brink@audrey.mpi-stuttgart.mpg.de

[1] H. A. Jahn and E. Teller, Proc. R. Soc. London, Ser. A **161**, 220 (1937).
[2] G. A. Gehring and K. A. Gehring, Rep. Prog. Phys. **38**, 1 (1975).
[3] K. I. Kugel' and D. I. Khomskii, Zh. Eksp. Teor. Fiz. **64**, 1429 (1973) [Sov. Phys. JETP **37**, 725 (1973)]; Usp. Fiz. Nauk **136**, 621 (1982) [Sov. Phys. Usp. **25**, 232 (1982)], and references therein.
[4] J. B. Goodenough, *Magnetism and Chemical Bond* (Interscience, New York, 1963).
[5] K. I. Kugel' and D. I. Khomskii, Fiz. Nizk. Temp. **6**, 207 (1980) [Sov. J. Low Temp. Phys. **6**, 99 (1980)].
[6] S. Ishihara, J. Inoue, and S. Maekawa, Physica C **263**, 130 (1996); Phys. Rev. B **55**, 8280 (1997).
[7] H. F. Pen, J. van den Brink, D. I. Khomskii, and G. A. Sawatzky, Phys. Rev. Lett. **78**, 1323 (1997).
[8] S. Marini and D. I. Khomskii, cond-mat/9703130 (unpublished).
[9] J. L. García-Muñoz, J. Rodríguez-Carvajal, and P. Lacorre, Europhys. Lett. **20**, 241 (1992).
[10] L. F. Feiner, A. M. Oles, and J. Zaanen, Phys. Rev. Lett. **78**, 2799 (1997).
[11] M. Wortis, Phys. Rev. **132**, 85 (1963).
[12] G. Khaliullin and V. Oudovenko, Phys. Rev. B **56**, 14 243 (1997).
[13] L. F. Feiner, A. M. Oles, and J. Zaanen (unpublished).