6

Spin-orbital Excitations

In this chapter the elementary excitations of a model Hamiltonian that captures the low energy behavior of a simple two-fold degenerate Hubbard Hamiltonian with Hund’s rule coupling, is studied. The phase diagram in the mean-field limit and in a two-particle approach reveals a rich variety of phases where both the orbital degrees of freedom and the spin degrees of freedom are ordered. We show that besides usual spin-waves (magnons) there exist also orbital-waves (orbitons) and, most interestingly, in a completely ferro-magnetically coupled system, a combined spin-orbital excitation which can be visualized as boundstates of magnons and orbitons. Both in one- and two-dimensions, for a completely degenerate system, the boundstates are found to be the lowest lying elementary excitations, much lower than the single spin or single orbital excitations. Finally we extend our treatment to almost-degenerate systems. The elementary excitations in orbitally degenerate strongly correlated electron systems in general carry both spin and orbital character.

6.1 Introduction

The \( d \) and \( f \) wavefunctions of free atoms are, besides Kramers (spin) degenerate, also a 5 and 7-fold orbital degenerate. If atoms with open \( d \) and \( f \) shells are collected into a solid, these orbitals retain to a large extend their atomic character because \( d \) and \( f \) wavefunctions are very localized around the nucleus. The potential for the electrons is mainly due this one atom and the overlap between orbitals on different atoms is small. This is the reason that in transition metal oxides the treatment of considering just one transition metal atom with nearest neighbor oxygen ligands is a fruitful starting point to describe the electron system, although translational

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symmetry is completely neglected in this approach. The ligand atoms, however, give rise to a correction to the potential of, say, a 3$d$ electron of the cation. This crystal-field potential in general lifts the 5-fold orbital degeneracy, but the degeneracy is not completely lifted if the crystal field has high enough symmetry. Typical examples of these so-called Jahn-Teller (J-T) ions are Cu$^{2+}$ ($d^9$) in an octahedral symmetric crystal field which has one hole in a two-fold $e_g$ orbital and Mn$^{3+}$ ($d^4$) and Cr$^{2+}$ ($d^4$) in octahedral symmetry (one hole in $e_g$). A lowering of the symmetry, for instance due to a lattice distortion can lift the remaining degeneracy and make the system gain energy [1]. If these strong Jahn-Teller ions are put as an impurity in a lattice their tendency to break the symmetry leads to lattice distortions. When Jahn-Teller ions are abundant and form a sub-lattice the corresponding lattice distortions are not independent. An interaction between them arises because the movement of a ligand ion towards a particular J-T center implies a movement away from another another J-T ion, stabilizing different orbitals of the two J-T centers. This makes the distortions cooperative and leads to structural phase transitions and a specific ordering of the orbitals. The J-T distortions are thus accompanied by a change in the space-charge distribution, which in turn causes a strong coupling to phonons.

Another important feature of transition metal oxides is their magnetism. So the partially filled 3$d$ level can have both a localized orbital and spin moment. In general these two moments are strongly coupled [2]. For these ions the coupling originates not from direct spin-orbit interaction as in heavy atoms, but via the conventional super-exchange, which in the case of orbital degeneracy couples not only to the spins, but also to the orbitals at neighboring sites. This is actually closely related to the well-known Goodenough-Kanamori-Anderson rules for exchange [3]. These rules state that if two electrons are in orbitals that directly overlap, their exchange is strong and anti-ferromagnetic and if they are in orbitals that do not directly overlap their exchange is weak and ferromagnetic. This can lead to situations where the spin (super-)exchange is maximized if the orbitals are ordered in a particular way. Recently the interplay between orbital ordering and magnetism has received a lot of attention, particularly in relation to the Colossal Magneto-resistance materials and Vanadium oxides [4-11].

Such a coupling between spin and orbital degrees of freedom, and in particular its influence on the excitation spectrum (in particular the collective modes) of such systems, is the topic of this chapter. To illustrate these ideas we take the two-fold degenerate Hubbard model with a Hunds-rule atomic exchange coupling as an example.

### 6.2 Degenerate Hubbard Model

The Hubbard Hamiltonian for a fully degenerate system consists of three parts: one for the hopping between orbitals on neighboring sites, one for the Coulomb repulsion for electrons on the same site (that are not necessarily in the same orbital) and one for the Hund’s rule exchange coupling between electrons on the same site, but in different orbitals:

$$H = H_t + H_U + H_J$$

(6.1)
with

\[ H_t = \sum_{<ij>,\sigma,\alpha,\beta} t_{ij}^{\alpha\beta} c_{i\alpha\sigma}^\dagger c_{j\beta\sigma} \]

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

\[ H_J = \sum_{i,\alpha,\beta} -J_R^{\alpha\beta} S_{i\alpha} \cdot S_{i\beta} (1 - \delta_{\alpha,\beta}), \]

where as usual \( n_m = c_m^\dagger c_m \) and \( S_m^+ = c_{m\uparrow}^\dagger c_{m\downarrow} \), \( S_m^- = c_{m\downarrow}^\dagger c_{m\uparrow} \) and \( S_m^z = \frac{1}{2}(n_{m\uparrow} - n_{m\downarrow}) \).

The indices \( \alpha, \beta \) denote orbitals, \( \sigma \) spin and \( <ij> \) nearest neighbor sites. There are a lot of parameters in the Hamiltonian, and we take \( J_R^{\alpha\beta} = J_H, U^{\alpha\beta} = U \) and \( t_{ij}^{\alpha\beta} = t_{ij} \) in order to simplify the Hamiltonian. The last simplification is quite drastic as it is well known that hopping integrals strongly depend on the relative orientation of the two orbitals. However, for our purposes, the investigation of the general structure of the excitation spectrum, it is sufficient to work with this simplified Hamiltonian. Consider a two-fold degenerate system with on average one electron per site in the strong coupling limit \( (U >> t) \). In this case we can integrate the doubly occupied states out, just as in deriving the Heisenberg Hamiltonian from the Hubbard Hamiltonian, and obtain in second order perturbation theory [12]

\[ H = \frac{2t^2}{U} (1 - \frac{J_H}{U}) \sum_{<ij>} S_i \cdot S_j + \frac{2t^2}{U} (1 + \frac{J_H}{U}) \sum_{<ij>} T_i \cdot T_j \]

\[ + \frac{2t^2}{U} (1 + \frac{J_H}{U}) \sum_{<ij>} (S_i \cdot S_j) (T_i \cdot T_j), \]

where the orbital operators are given by:

\[ T_i^+ = c_{i\uparrow\beta}^\dagger c_{i\uparrow\alpha} + c_{i\downarrow\beta}^\dagger c_{i\downarrow\alpha} \]

\[ T_i^- = c_{i\uparrow\alpha}^\dagger c_{i\uparrow\beta} + c_{i\downarrow\alpha}^\dagger c_{i\downarrow\beta} \]

\[ T_i^z = \frac{1}{2} (n_{i\uparrow\beta} + n_{i\downarrow\beta} - n_{i\uparrow\alpha} - n_{i\downarrow\alpha}) \]

The commutation relations for the orbital operators are exactly the same as for spin \( \frac{1}{2} \) operators, so that we can refer to the orbital operators as pseudo-spin operators. The spin and pseudo-spin operators commute. Note that the relative simple form of the Hamiltonian above is mainly due to the simplifications made to the hopping integrals. In general also more involved combinations of spin and orbital operators turn up in the Hamiltonian [2, 13].

In the Hamiltonian 6.5 the spin degrees of freedom are coupled to the orbital degrees of freedom and ordering of orbitals couples to ordering of the spins. This is illustrated in figure 6.1 where the exchange between two spins on different sites is shown for the non-degenerate and two-fold degenerate situation. In the former case spins tend to be aligned anti-ferromagnetically, as opposed to the latter situation.
where spins prefer to be in different orbitals and to be aligned ferromagnetically, corresponding to a ferromagnetic spin alignment and an "anti-ferromagnetic" orbital alignment. The strong intra-atomic Hund’s rule exchange coupling is the principal interaction behind this effect.

### 6.3 Spin-Orbital Hamiltonian

The effective exchange Hamiltonian for materials containing (almost) degenerate localized electron levels and spin degrees of freedom depends on the overlap integrals between the localized orbitals and is, in general, quite complex. However, the main feature of the system under study is the presence of two interacting spin variables, and the qualitative behavior of such systems can be captured in a model Hamiltonian of the form:

\[
H = -J_s \sum_{<ij>} \mathbf{S}_i \cdot \mathbf{S}_j - J_t \sum_{<ij>} \mathbf{T}_i \cdot \mathbf{T}_j - 4J_{st} \sum_{<ij>} \left( \mathbf{S}_i \cdot \mathbf{S}_j \right) \left( \mathbf{T}_i \cdot \mathbf{T}_j \right),
\]

where \( \mathbf{S}_i \) and \( \mathbf{T}_i \) are the spin and pseudo-spin (orbital) operators and \( <ij> \) denotes a summation over nearest neighbors. Note that terms of the form \( \mathbf{S} \cdot \mathbf{T} \), that describe spin-orbit coupling, are not present in this Hamiltonian. The coupling between spin and orbital degrees of freedom, given in the last part of equation 6.7, arises from the Goodenough-Kanamori-Anderson rules for exchange: the size and sign of the exchange coupling between two spins depends on the overlap of their electron-wavefunctions [3]. This class of models has been studied in the context of magnetic materials with orbital degeneracy [2,14], but also has broader relevance, for example, to the description of charge and spin ordering in quasi one-dimensional organic salts [15].
6.4 Groundstate

If our aim is to calculate the elementary excitations of a system described by the Hamiltonian 6.7, we first have to know the groundstate wavefunctions. We can easily calculate a phase diagram within two different approximations: the mean-field and the two spin approximation. In the latter only two interacting spins and pseudo-spins are considered, so that the groundstate is described in a way similar to a valence-bond state, which is a good starting point for low dimensional spin systems. Starting from groundstate wavefunctions obtained within one of these approximations, one can test the stability of the groundstate by calculating its response to for instance spin-waves and combined orbital- and spin-waves.

6.4.1 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

\[ H_{s}^{MF} = -J_{s} \sum_{<ij>} \mathbf{S}_{i} \cdot \mathbf{S}_{j} - 4J_{st} \sum_{<ij>} < \mathbf{T}_{i} \cdot \mathbf{T}_{j} > \mathbf{S}_{i} \cdot \mathbf{S}_{j} \]

\[ = -J'_{s} \sum_{<ij>} \mathbf{S}_{i} \cdot \mathbf{S}_{j} \quad (6.8) \]
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Figure 6.3: Phase-diagram of the two-particle solution of the model Hamiltonian. In the 11 phase both spin and pseudo-spin are in a triplet state, in the phase 10 the spin is triplet, pseudo-spin singlet, in the phase 01 the spin is singlet, pseudo-spin triplet and in the phase 00 both spin and pseudo-spin are in a singlet state. $J_s$ and $J_t$ are in units of $|J_{st}|$.

and

$$H_{t}^{MF} = -J_t \sum_{<ij>} T_i \cdot T_j - 4 J_{st} \sum_{<ij>} <S_i \cdot S_j> T_i \cdot T_j$$

$$= -J_t' \sum_{<ij>} T_i \cdot T_j.$$

In Hamiltonian 6.8 the orbital degrees of freedom are integrated out and in Hamiltonian 6.9 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 (as for instance done in [9]) we have, in some sense, thrown away the interesting part of our problem and returned to a renormalized Heisenberg model, where groundstate properties and elementary excitations are well known. Nevertheless, as in a first approximation it is still a useful approach, the phase diagram is given in figure 6.2.

6.4.2 Two-Spin Solution

In mean-field short range interactions are averaged out, so that if we want to gain insight in local properties it is more useful to break the translational symmetry and only consider a few interacting particles. In this case we consider two spins and two pseudo-spins and obtain the Hamiltonian

$$H_{1,2} = -J_s S_1 \cdot S_2 - J_t T_1 \cdot T_2 - 4 J_{st} (S_1 \cdot S_2) (T_1 \cdot T_2).$$

The combinations of (pseudo-)spin operators can either be singlet or triplet and the groundstate energy of the various combinations is shown in the table below and the corresponding phase diagram in the figure 6.3, for $J_{st} > 0$ and $J_{st} < 0$, respectively.
Figure 6.4: Character of the first excited state in the two-spin system. $J_s$ and $J_t$ are in units of $|J_{st}|$.

The character of the first excited state for the various phases for $J_{st} > 0$ is shown in figure 6.4. 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 spin approach. In the white, central part of this figure the groundstate 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 the excitations of a system with such an interaction carry both spin and orbital character.

### 6.5 Ferro-Ferro system

When $J_s$, $J_t$ and $J_{st}$ are chosen in a range where both the spins and pseudo-spins order ferromagnetically, a ferro-ferro phase, we can obtain exact analytical expressions for the elementary excitations. In a ferromagnetic Heisenberg model the exact groundstate is the state with all spins pointing in the same direction, as opposed to a Heisenberg Hamiltonian with an anti-ferromagnetic exchange where quantum fluctuations affect the Neél spin order.
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Figure 6.5: Excitation energy for Ising spins and pseudo-spins for the ferro-ferro system. When the spin and orbital are close to together, the excitation energy is lower than in the case they are far apart.

The coupling between the spins and pseudo-spins can cause the elementary excitations of the system to be bound states of spin and pseudo-spin excitations. This is illustrated for the ferro-ferro system in figure 6.5. Let us consider Ising spins (and pseudo-spins) and examine the energy of an excitation of one spin plus one pseudo-spin. If the two excited spins are far away from each other, the excitation energy due to the spin pseudo-spin interaction is \(8|J_{st}|\). When the excited spins are close to each other, on the nearest neighbour sites or on the same site, the excitation energy is \(4|J_{st}|\) and 0, respectively. This means that a magnon and an orbiton (orbital wave) have an attractive interaction and that, depending on dimensionality and strength of the attraction, the spinon and orbiton can form a boundstate.

6.5.1 Equations of motion

In order to establish whether it is possible to obtain bound states that are combinations of orbital- and spin-waves in the excitation spectrum of the model system, we will use the equation of motion method, which provides a simple, and in the ferromagnetic case exact, way to calculate boundstate energies. Before addressing this issue, let us first examine a single (pseudo-)spin excitations. Starting from Hamiltonian 6.7 and with a groundstate \(|0\rangle\) 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:

\[
(H - E_0)S_m^- |0\rangle + \omega_s S_m^- |0\rangle = [H, S_m^-] |0\rangle.
\] (6.11)

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

\[
\omega_s(Q) = 2(J_s + J_{st}) \sum_a (1 - \cos Q \cdot a)
\] (6.12)
and

\[ \omega_a(Q) = 2(J_t + J_{st}) \sum_a (1 - \cos Q \cdot a), \]

(6.13)

where the lattice vectors are denoted by \( a \). The presence of the coupling between the spin and orbital degree of freedom, parameterized by \( J_{st} \), thus merely renormalizes the spin/orbital wave spectrum.

The equation of motion for a combined spin and pseudo-spin excitation \( S_m^{-} T_n^- \) is

\[ (H - E_\omega)S_m^- T_n^- \ket{0} = \omega S_m^- T_n^- \ket{0} = \{ H, S_m^- T_n^- \} \ket{0}. \]

(6.14)

We find for the commutator

\[
[H, S_m^{-} T_n^-] = -(J_s + J_{st}) \sum_a (S_m^- - S_{m+a}^-) T_n^- \\
- (J_t + J_{st}) \sum_a (T_n^- - T_{n+a}^-) S_m^- \\
- 2J_{st} \sum_a (\delta_{m,n} - \delta_{m,n+a})(S_m^- - S_{m+a}^-)(T_n^- - T_{n+a}^-),
\]

where the lattice vectors are denoted by \( a \). The Fourier transform of the equation of motion is

\[
\{ \omega - \gamma(Q, q) \} \cdot A(Q, q) = -8J_{st} \left( \frac{a}{2\pi} \right)^d \sum_a (\cos Q \cdot a/2 - \cos q \cdot a) \\
\cdot \int d\mathbf{k} (\cos \mathbf{Q} \cdot \mathbf{a}/2 - \cos \mathbf{k} \cdot \mathbf{a}) A(Q, \mathbf{k}),
\]

(6.16)

with

\[ A(Q, q) = S_{Q/2-q}^- T_{Q/2+q}^- \ket{0} \]

(6.17)

\[ \gamma(Q, q) = \epsilon_s(Q/2 + q) + \epsilon_t(Q/2 - q) \]

(6.18)

\[ \epsilon_s(k) = 2(J_s + J_{st}) \sum_a (1 - \cos k \cdot a) \]

(6.19)

\[ \epsilon_t(k) = 2(J_t + J_{st}) \sum_a (1 - \cos k \cdot a) \]

(6.20)

and \( d \) the dimensionality of the system. The total momentum of the excitation is \( Q \) and the relative momentum of the combined spin and pseudo-spin excitation is \( \mathbf{k} \). In order to check if there is a self-consistent solution of the equation of motion, equation 6.16 is iterated once. Then a set of equations is found that can be represented
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in a matrix \(|M_{\alpha,\beta}|\) of order \(d\) for hyper-cubic lattices. In a three-dimensional system \(\alpha, \beta = x, y, z\). The set of equations has a solution if, and only if,

\[ \text{Det} \ |\delta_{\alpha,\beta} - M_{\alpha,\beta}| = 0 \]  \hspace{1cm} (6.21)

with,

\[ M_{\alpha,\beta}(Q) = -8J_{st} \left( \frac{a}{2\pi} \right)^d \]
\[ \cdot \int d\mathbf{k} \frac{(\cos Q_{\alpha}/2 - \cos k_{\alpha})(\cos Q_{\beta}/2 - \cos k_{\beta})}{\omega - \gamma(Q, \mathbf{k})} \]  \hspace{1cm} (6.22)

If the system is one-dimensional the condition above reduces to

\[ 1 = -\frac{8J_{st}}{2\pi} \int_{-\pi}^{\pi} dk \frac{(\cos Q/2 - \cos k)^2}{\omega - \epsilon_s(k + Q/2) - \epsilon_t(k - Q/2)}, \]  \hspace{1cm} (6.23)

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 [16].

6.5.2 Boundstates

We restrict ourselves to the ferro-ferro phase, i.e. according to figure 6.3 \(2J_{st} < J_s + J_t\) and \(J_{st} > 0\), and determine the boundstate energies from condition 6.21 and 6.22. 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 \(\omega_c(Q)\) where the denominator of the matrix elements \(M_{\alpha,\beta}\) start to diverge. The condition for this is:

\[ \omega_c(Q) = \text{Min}_k[\epsilon_s(Q/2 + k) + \epsilon_t(Q/2 - k)] \]  \hspace{1cm} (6.24)

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

\[ \omega_s(Q) \geq \omega_c(Q) \]
\[ \omega_o(Q) \geq \omega_c(Q). \]

The explanation for this is that in a combined spin- and orbital-excitation the momentum is distributed over the two sub-systems and because the excitation spectrum is non-linear (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 will consider the one-dimensional case. The integral in equation 6.23 can be reduced to the form

\[ \frac{(a - \cos k)^2}{1 + b \cos k + c \sin k}, \]  \hspace{1cm} (6.25)
which is known analytically. The condition that the integral be equal to unity then results in finding the roots of a third order polynomial. Not so much is gained following this procedure, as the resulting expressions are very long and complex. So let us consider some special momenta in the Brillouin zone.

- **Q = 0**

  In the one-dimensional case, for $Q = 0$ the condition 6.23 for a boundstate at $\omega = 0$ reduces to: $2J_{st} = J_s + J_t$, which is exactly at the phase boundary of the ferro-ferro phase. A bound state at negative $\omega$ appears when $2J_{st} > J_s + J_t$. This simply means that the groundstate is not stable (by exciting it energy is gained), which we expected as the phase diagram shows that the anti-ferro anti-ferro phase is the groundstate in this parameter range.

- **Q = $\pi$**

  By treating this special case, we can prove that there is always a boundstate in the ferro-ferro system, at least in one-dimension. When $Q_{\alpha} = \pi$ for all $\alpha$, at the corner of the Brillouin zone, the equations simplify considerably. The off-diagonal matrix elements in Eq. 6.21 vanish so that $M_{\alpha,\beta} = D \delta_{\alpha,\beta}$ in any dimension. This yields

  $d=1$

  \[
  \omega_{b}^{ld}(\pi) = 2(J_s + J_t) - (J_s - J_t)^2/4J_{st} \\
  \omega_{c}^{ld}(\pi) = 4(\min|J_s, J_t| + J_{st}) \\
  \]

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

- **At $\omega(Q) \rightarrow \omega_{c}(Q)$**

  It follows from equation 6.23 that for $\omega \rightarrow -\infty$, $M_{\alpha,\alpha}(Q) \rightarrow 0$, so that there can never be a boundstate for large negative $\omega$. For $\omega(Q) \rightarrow \omega_{c}(Q)$, (for energies approaching the continuum) the integrand in equation 6.23, except for $Q = 0$, diverges as $k^{-1}$, making the integral logarithmically divergent. We can conclude from this that for any $Q$ 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 $0 \leq Q \leq \pi$ a spin-orbital boundstate develops and that this boundstate, as it is by definition lower in energy than the spin-orbital continuum, which in turn, in a certain parameter range is lower than the single (pseudo-)spin excitations, is the lowest lying, elementary excitation of the ferro-ferro system in this range. Before illustrating the statement above with numerical examples, let us consider one more special case, namely:
Figure 6.6: Dispersion of the spin-orbital boundstate, spin-orbital continuum, 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 $J = J_s$.

- $J_s = J_t = J_{st}$ in 1d
  For these parameters the system is exactly at the phase boundary of the ferro-ferro groundstate. Now equation 6.23 takes a particular simple form, and the solution for the boundstate is
  \[ \omega_b^*(Q) = 2J_{st}(1 - \cos Q). \]  
  \hspace{1cm} (6.28)

  For the lower bound of the continuum, the single-spin and the single-orbital excitations one obtains in this case
  \[ \omega_o^*(Q) = 8J_{st}(1 - \cos Q/2), \]  
  \[ \omega_o^*(Q) = \omega_o^*(Q) = 4J_{st}(1 - \cos Q). \]  
  \hspace{1cm} (6.29) \hspace{1cm} (6.30)

  In figure 6.6 the dispersion relations of these excitations are shown. The spin-orbital boundstate is always the lowest energy excitation of the system. Note that for small $Q$ the spin-orbital continuum and the boundstate are very close in energy and their energy difference is of the order of $\frac{J_{st}}{16} Q^1$.

  The numerical solution of the boundstate equation for a one-dimensional system is shown in figure 6.7 for two parameter sets. For small momenta the boundstate 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 boundstate 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 orbital excitation spectra all coincide.
Figure 6.7: Dispersion of the spin-orbital boundstate, spin-orbital continuum, single spin and single orbital excitations in a one-dimensional ferro-ferro system. The unit of energy is $J = J_s$.

In the right part of figure 6.7 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 boundstate, continuum and magnon excitations are close in energy, and in the limit that $J_t \to \infty$ all three of them coincide.

In figure 6.8 a typical result for a two dimensional system is shown. It is found numerically that there always exists at least one boundstate, also in two dimensions. Similar to the 1D case, the boundstate is only well separated from the continuum at the edge of the Brillouin-zone. If $J_s = J_t$ it can be shown that

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

and

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

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

### 6.5.3 Almost Degeneracy

In the sub-sections 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 $\alpha$ and $\beta$ is $\Delta$. The Hamiltonian to be added to equation 6.1 is

$$H_\Delta = \Delta/2 \sum_{i, \sigma} (n_{i, \sigma, \beta} - n_{i, \sigma, \alpha})$$

$$= \Delta \sum_i T_i^\sigma,$$

(6.33)
where the last equality follows from equation 6.6. The level-splitting manifests itself in the pseudo-spin language as a magnetic field for the orbitals. Carrying through the calculation for the excitations leads to renormalized spin-waves, orbital-waves and boundstates:

\[ \bar{\omega}_s = \omega_s \]
\[ \bar{\omega}_t = \omega_t + \Delta \]
\[ \bar{\omega}_{st} = \omega_{st} + \Delta. \] (6.34)

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 boundstate energy being gapped for the same reason. The equalities above permit a convenient generalization of the results derived for the fully degenerate system to the situation where the levels are non- (or almost-) degenerate, as illustrated in figure 6.9, where the dispersion of the boundstate in the case of an orbital energy splitting of \( \Delta = 2J_s \) is shown.

### 6.6 Conclusions

In this chapter a model Hamiltonian that captures the low energy behavior of a two-fold degenerate Hubbard Hamiltonian with Hund’s rule coupling, was studied. We presented the phase diagram in the mean-field limit and in a two-particle approach, revealing a rich variety of phases where both the orbital degrees of freedom and the
spin degrees of freedom can be ordered (anti-)ferromagnetically. We have shown that in this case there may exist, besides usual spin-waves (magnons) also orbital-waves (orbitons) and, most interestingly, the combined spin-orbital excitation which can be visualized as boundstates of magnons and orbitons. Both in one- and two-dimensions the boundstates are found to be the lowest lying elementary excitations, much lower than the single spin or single orbital excitations. This shows that the elementary excitations in orbitally degenerate strongly correlated electron systems in general carry both spin and orbital character.

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