SPIN AND ORBITAL EXCITATIONS ON FINITE CHAIN

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Abstract

The finite chain of singly occupied twofold degenerate orbital sites with Hund’s rule coupling is considered. The equation of motion for the combined spin and orbital excitation is solved exactly. The structure and stability of bound versus scattered states is examined and the critical momenta for state crossing are determined. The solutions are also extended to the thermodynamic limit of an infinite chain and compared to the existing spin-orbiton descriptions. The possibility to probe orbiton dynamics via spin excitations is discussed.

PACS numbers: 75.30.-m, 75.30.Et, 75.40.Gb, 75.10.Jm

1. INTRODUCTION

Orbital degrees of freedom are especially important for the systems containing transition metal ions with degenerate or pseudo-degenerate strongly correlated energy levels such as cuprates, manganates, vanadates etc.. Besides of the interest related to the emerging new materials and their applications, these systems have attracted a large interest for their fundamental properties. Ordering and excitations of charge, spin, orbital and other degrees of freedom in such systems are deeply interconnected giving rise to a variety of new properties (see [1] for a review). A recent example is the experimental observation of the orbiton, a fundamental excitation in a solid with long range ordering of electron orbitals, LaMnO$_3$ [2]. The low energy excitations can be described by means of effective models after projecting on the subspace of a relevant energy scale, e.g. [3]. Generally, the coupling constants in such models are anisotropic and antiferro ordering of one subsystem favours ferro ordering in the other one. However there can exist situations when both subsystems order ferromagnetically. These can be associated with phase separated or stripe phases observed at certain doping in manganites with colossal magnetoresistance like La$_{1-x}$Ca$_x$MnO$_3$ or in transition metal oxides like NaNiO$_2$ with a layered frustrated lattice. A completely ferromagnetically coupled system was considered in [4], where a new type of composite, spin and orbital, bound excitation was described in the thermodynamic limit on 1$D$ and 2$D$ lattices. Bound excitations, as being the lowest in energy, can be very important for the low temperature response of the system, for the formation of inhomogeneous phases and others. In the present paper we describe an exact solution of this model for a finite chain. It allows us to get a deeper insight into the nature of the spin-orbiton excitation. It turns out that to understand the specific features of the bound excitation it is important to analyze the spectra of the scattering states, which are difficult to observe within the thermodynamic limit treatment itself.

2. THE EQUATION OF MOTION

We consider the following Hamiltonian describing the low energy spin and orbital excitations of electrons in doubly degenerate orbital states:

\[
H = -\sum_{i=0}^{N-1} \left( J_S S_i \cdot S_{i+1} + J_O T_i \cdot T_{i+1} + 4J_{SO} (S_i \cdot S_{i+1})(T_i \cdot T_{i+1}) \right)
\]  

(1)

Qualitative arguments and the energy spectrum were given to prove the existence of the combined bound state for arbitrary interaction strength [4] in analogy with the two-magnon excitation in a simple ferromagnet [5]. This bound state was found to be the lowest energy elementary excitation of the system. We show that in the thermodynamic limit there exist two combined bound state modes (ST) which differ by symmetry of the wave function but have the same dispersion. However these are not the lowest energy excitations: for any choice of parameters corresponding to the ferromagnetic ground state either the two-magnon (SS) or the two-orbiton (TT), or both of them have
a lower excitation energy at any momenta. Moreover, if magnons and orbitons have a different stiffness, i.e., \( x = J_x - J_y \neq 0 \), there exists a critical value of \( x \), beyond which the ST bound state disappears at the edge of the Brillouin zone, \( P = \pi \). For a finite lattice the instability of the ST excitation becomes even more dramatic. The spectrum splits into two branches and both bound states merge with the continuum of scattered states in finite areas close to the \( P = 0 \) and \( P = \pi \). This behavior is in contrast with the pure SS or TT excitations, where a stable bound state exists for arbitrary momenta.

The spin-orbiton excitation is defined in the usual way

\[
| \psi > = \sum_{0 \leq n_1, n_2 < N - 1} a_{ST}(n_1, n_2) S_{n_1}^{-} T_{n_2}^{-} | 0 > .
\]

(2)

By separating the total momentum \( P \) we introduce the amplitude of the relative distance \( X = n_2 - n_1 \) between spin and orbital flips

\[
a_{ST}(n_1, n_2) = \exp \left( iP \frac{n_1 + n_2}{2} \right) A(X),
\]

(3)

which is decomposed in the Fourier series

\[
A(X) = \frac{1}{\sqrt{N}} \sum_{Q} \exp(iQX) B(Q).
\]

(4)

From the periodic boundary conditions \( a_{ST}(n_1, n_2) = a_{ST}(n_1 + N, n_2 + N) \) and \( a_{ST}(n_1, n_2) = a_{ST}(n_1, n_2 + N) = a_{ST}(n_1, n_2 + N) \), one obtains the quantization of the total momentum \( P = 2\pi k/N, \quad k = 0, 1, \ldots, N - 1 \) and a relation satisfied by the amplitude of relative motion

\[
A(X) = \exp(i\pi k) A(X + N)
\]

\[
A(X) = \exp(i\pi k) A(N - X).
\]

(5)

Equation (5) allows to classify the states into symmetric (s) and antisymmetric (a), depending on even or odd values of \( k \). As a consequence of this relation one obtains two sequences of values for \( Q \) in the Fourier expansion (4): \( Q^s = 2\pi m/N \) or \( Q^a = 2\pi \left( m + \frac{1}{2} \right)/N \), \( m = 0, 1, \ldots, N - 1 \). Equation (6) in a similar way determines the three sequences depending on the values of \( k \): \( k_1 = 0, 3, 6, \ldots; k_2 = \ldots \)
$1,4,7,\ldots; k_3 = 2,5,8,\ldots; 1 2$

These sequences correspond to three distinct modes which can exist in the three magnon sector of the excitation. Returning to the two-magnon sector and taking the Fourier transform of the equation of motion we obtain

$$B(Q) \left[ E - (J_S + 1) \left( 1 - \cos \left( \frac{P}{2} - Q \right) \right) - (J_T + 1) \left( 1 - \cos \left( \frac{P}{2} + Q \right) \right) \right]$$

$$= -4 \left[ \cos \left( \frac{P}{2} \right) - \cos (Q) \right] \frac{1}{N} \sum_Q B(Q) \left[ \cos \left( \frac{P}{2} \right) - \cos Q \right],$$

(7)

where the excitation energy $E$ and the coupling constants are scaled with the spin-orbital coupling constant $E \rightarrow E(J_{ST})$, $J_s \rightarrow J_s(J_{ST})$, $J_T \rightarrow J_T(J_{ST})$. For a fixed momentum $P$ the amplitude is

$$B(Q) = \frac{C \cos \delta}{\cos (\frac{P}{2}) - \cos (Q)} \times \frac{\cos (\frac{P}{2}) - \cos Q}{\cosh \nu - \cos (Q - \delta)},$$

(8)

where we have introduced two phase variables:

$$\delta = \arctan \left( \tan \left( \frac{P}{2} \right) \right) \left( J_s - J_T \right)$$

$$v = \arccosh \left( \frac{\cos (\delta)}{\cos (\frac{P}{2})} \left( 1 - \frac{E}{2 + J_s + J_T} \right) \right),$$

(9)

Iterating the definition of the constant $C = \frac{1}{N} \sum_Q B(Q) \left[ \cos (\frac{P}{2}) - \cos Q \right]$ leads to the eigenenergy equation

$$1 = \frac{4 \cos (\delta)}{N \left( 2 + J_s + J_T \right) \cos (\frac{P}{2})} \sum_{m=0}^{N-1} \left( \cos \left( \frac{P}{2} \right) - \cos Q_m \right)^2.$$ (10)

The integral form of this equation for $N \rightarrow \infty$ coincides with the result in [4] up to a factor of 2 used to scale the coupling constants. The meaning of introducing the variables in (9) becomes now clear: bound states, if any, correspond to real valued positive solution for $\nu$ in (10) while scattered states, which are related to the singularities of the denominator, correspond to purely imaginary values of $\nu$. The threshold value $\nu = 0$ establishes a separation line between the two types of solutions:

$$E_c = 2 + J_s + J_T - \sqrt{4 \cos^2 \left( \frac{P}{2} \right) \left( 1 + J_T \right) \left( 1 + J_s \right) + \left( J_s - J_T \right)^2}.$$ (11)

The phase $\delta$ quantifies the difference of dynamic properties of magnon and orbiton excitations throughout the Brillouin zone. The values of $\delta \in [0, \pi/2]$ and increase from the center towards the edge.

3. EXACT SOLUTION FOR THE FINITE CHAIN

To solve Eq.(7) we use the following general expressions for finite sums (details of derivation to be given elsewhere):

$$\sinh \nu \sum_{m=0}^{N-1} \frac{\sin \left( \frac{Q}{N} + 2\pi \frac{Q}{N} X \right)}{\cosh \nu - \cos \left( \frac{Q}{N} + \delta + 2\pi \frac{Q}{N} \right)}$$

$$\frac{108}{N}$$
\[
\sinh v \sum_{n=0}^{N-1} \frac{\cos \left( \left( \frac{\pi}{N} + \frac{\pi}{2} \right) X \right)}{\cosh v - \cos \left( \frac{\pi}{N} + \frac{\pi}{2} + 2\pi \frac{n}{N} \right)}
\]

(12)

where the phase variables can take complex values and \(X \in [0, N-1]\). Extension outside the physical interval is described by (5). For instance, one can check that with (12) Bethe’s solution for a spin chain is recovered at once without making an ansatz. We note that for the considered system the solution of Bethe’s problem describes either the two magnon or two orbiton excitation with renormalized exchange interactions:

\[
J = J_S + J_{ST} \quad \text{and} \quad J = J_T + J_{ST}
\]

respectively. The values of \(\theta\) for the combined ST excitation are fixed by the condition \(\theta = PN\) in (12). As a result we obtain the following equation for the eigenenergy, irrespective of the symmetry of the mode:

\[
\left( J_S + J_T - 2 \right) \cos \left( \frac{\theta}{2} \right) \sinh \left( v \right) = \frac{4 \cos \left( \delta \right) \sinh \left( v \right)}{\left( \cos \left( \frac{P}{2} \right) - e^{-\nu} \cos \left( \delta \right) \right) \left( \cos \left( \frac{P}{2} \right) - e^{\nu} \cos \left( \delta \right) \right) - \frac{\sinh \left( v \right)}{\cos \left( \delta \right)}}
\]

(13)

One can notice that the wildly oscillating terms containing \((\delta N)\) in the last two lines of the equation are suppressed in thermodynamic limit, provided we are considering the bound states (real and positive values of \(v\)). In the simplest case, \(J_S = J_T\), the behavior of the ST states is qualitatively similar to that of SS or TT excitations and coincides with Bethe’s solution when \(J_{ST} = J = J_{ST}\). For instance, one finds that a stable bound state exists for an arbitrary magnitude of coupling constants at any finite momenta \(P\). As in Bethe’s solution, the antisymmetric bound state becomes unstable and decays into scattering states in the long wavelength region of the Brillouin zone which scales as \(1/\sqrt{N}\). However, as soon as \(J_S\) and \(J_T\) become different, the distinctive features of the ST excitations begin to emerge. Both symmetric and antisymmetric bound states become unstable at small momenta \(P\), i.e., they cross the boundary of scattering states at some \(P = P_c \neq 1\). Let us consider the symmetric excitation. Solving Eq.(13) for \(v(P_c) = 0\) leads to

\[
P_c^4 = \frac{2}{N} \left( J_S + J_T - 2 \right) \sin^2 \left( \frac{J_S - J_T}{2} \times \frac{PN}{4} \right) \left( \frac{2 + J_S + J_T}{(J_S + 1)(J_T + 1)} \right)^2 + O\left( \frac{1}{N^2} \right).
\]

(14)

Not only the symmetric state becomes unstable, but also the region of instability has become discontinuous due to the oscillating term. But most unexpectedly, an instability appears for short
wavelengths of the excitation, where one would normally expect the strongest binding and localization to take place. Indeed, for $|J_S - J_T| > 4J_{ST}$ a rather broad instability region is found from Eq.(13) close to the edge of the Brillouin zone $(\pi - P) \ll 1$:

$$P_c = \pi - \sqrt{\frac{J_S - J_T - 4}{2N}} \frac{J_S - J_T}{J_S + 1} \sin \left( \frac{P_c N}{4} \right) + O \left( \frac{1}{N} \right),$$

(15)

where we have assumed that $J_S > J_T + 4$. If, alternatively, $J_T > J_S + 4$, then the indices $S$ and $T$ have to be interchanged. The same equations hold for the antisymmetric excitation by replacing "$\sin$" with "$\cos$" in (14) and (15). We note that the long wavelength critical point has moved to shorter momenta $P_c \pi^{-1/4}$ as compared to Bethe’s spin chain $P_c \pi^{-1/2}$, but most remarkably, the above equation contains multiple solutions for $P_c$ for any finite $J_S \neq J_T$ because of the oscillating terms on the r.h.s. of the equations. This means that in the critical regions of the Brillouin zone which scale as $N^{-1/4}$ close to $P = 0$ and $N^{-1/2}$ close to $P = \pi$ one has to observe reentrant behavior of bound states. This behavior is a consequence of the radical change of the structure of the scattering bands. Due to the energy splittings at the crossings of non-interacting bands, the spectrum of the interacting bands acquires a layered structure with undulations within the layers. As a result, the lowest energy dispersion curve is formed which corresponds to the "reentrant" bound. The difference between symmetric and antisymmetric states, which is very significant for purely spin excitations [7], is almost completely wiped out for the combined excitations. The period of the undulations of the ST bands is determined by $\delta$ and $N$. It decreases towards small momenta, where the bands become more similar to the monotonous SS bands (see, e.g., [6]) if $\delta$ and $N$ are small enough. At first sight, the short wavelength behavior seems to be in contradiction with the physical arguments and calculations presented in [4]. For instance, the short wavelength instability of the bound state "survives" even in the thermodynamic limit. If the stiffness of orbitons is close to that of magnons one indeed finds a stable bound state for the whole Brillouin zone in analogy with the two magnon problem [4]. However, at larger values the spin-orbiton bound state disappears at the edge of the Brillouin zone despite the Goldstone mode singularity keeping its energy away from merging with continuum. Indeed, from the definitions (9) we have $\delta \rightarrow \pi/2$ and

$$\frac{\cos(\delta)}{\cos(\zeta)}_{P = \pi} = \frac{|J_T - J_S|}{2 + J_S + J_T}.$$

After substitution into Eq.(13) we obtain a simple relation:

$$\frac{|J_T - J_S|}{4} = e^{-\nu}.$$

(16)

Since for a bound state $\nu$ should be real and positive, this equation does not have solutions for $|J_S - J_T| > 4$. At the critical point the energy of the bound state reaches the lower boundary of the band of scattered states $E_c$, Eq.(11). Above this threshold Eq.(16) is not valid anymore since becomes purely imaginary and description of the band of scattering states requires knowledge of finite size corrections contained in Eq.(13). Thus, for the ST bound state exists at the intermediate momenta outside the critical regions described by Eqs.(14) and (15) and its energy almost coincides with the lower boundary of scattered states. These features demonstrate that the binding of mixed spin and orbiton excitations can actually be very weak even if the ST coupling is significant.

To understand the physical reason of these instabilities it is necessary to consider the behavior of the ST wave function on a finite chain. On one side, there is indeed an energy gain for the spin and orbital excitations to occur within the range of the ST coupling as explained in [4]. Consequently, one expects that, just as in the spin-spin problem, a tightly bound spin-pseudospin soliton is formed which moves as a single entity over the lattice. The larger the number of excited spins, the larger is
the energy gain of having droplets of such excitations. This is essentially the mechanism of domain wall formation which clearly remains valid for the spin-orbiton system. One obtains a stable bound state for any momenta which behaves similarly to the one found by Bethe. However, the situation changes qualitatively when. This is reflected in the explicit form of the wave function following from expressions (12).

\[
A(X) \propto \exp \left( \frac{i\delta}{2} \left( X - \frac{N}{2} \right) \right) \left( \frac{\exp \left( v \left( \frac{X}{2} - X \right) \right)}{\sinh \left( N \left( v + i \left( P + \delta \right) / 2 \right) \right)} + \frac{\exp \left( -v \left( \frac{X}{2} - X \right) \right)}{\sinh \left( N \left( v - i \left( P + \delta \right) / 2 \right) \right)} \right).
\]

First we note that a difference in the stiffness of the excitations means that the pseudo-particles move over the lattice with different velocity. From Eqs.(8-10) it also follows that \( \delta \) is a measure of momentum exchanged between the two interacting pseudoparticles and it becomes non-zero as soon as \( J_S - J_T \neq 0 \). This produces an oscillating factor \( \exp(iX\delta) \) in the wave function. At short wavelengths we have \( \delta \propto \frac{\pi}{2} \) and the wave function changes sign on the distance of a lattice spacing.

The dynamic potential represented by the variable \( \delta \) should be strong enough to make the binding possible. For the spin-spin problem, for instance, this is achieved due to the divergence of \( v \) at \( P = \pi, \) which leads to a strongly bound state of two spin deviations localized on strictly nearest neighbor sites. But the dynamic potential itself depends on the difference \( J_S - J_T \) and, as follows from (16), decreases very fast until reaching zero at the critical value. Diminishing \( v \) leads to a more delocalized shape of the wave function which allows the oscillating term to annihilate the effect of the attractive potential. This results in formation of a scattering or a resonance state. Away from the edge of the Brillouin zone the phase angle \( \delta \) becomes smaller and the bound state is stabilized at intermediate. However the competition of binding, \( v \), and unbinding, \( \delta \), tendencies determines the reentrant behavior in the critical regions, Eqs.(14) and (15). At longer wavelengths the bound state becomes unstable again due to the large extent of the wave function which has nodes \( \delta \neq 0 \) even for the symmetric. In a 2D system the same physical mechanisms are operative, but the critical regions are governed by logarithmic terms in \( N \) instead of power law dependences [7]. Therefore one has to expect that bound spin and orbital excitations would exist in a much more restricted region of the Brillouin zone at intermediate momenta. In a 3D system bound states are likely not to appear at all even in the thermodynamic limit.

Another distinctive property of the ST excitations is their stronger ability to destroy long range ordering in the system. Coupling of magnons and orbitons due to the biquadratic term in the Hamiltonian (1) acts to diminish the energy of the combined excitation. This leads to a stronger restriction such excitations set on the stability of the ground state, as compared to pure spin or orbital excitations. The restriction requires that excitation energy be positive. Keeping only the main terms in (13) for the \( P << 1 \) expansion we find in the limit \( N \to \infty. \)

\[
\frac{(J_S + J_T - 2)\sinh(v)}{4} = \left( \frac{E}{2 + J_S + J_T} - v \right) \left( \frac{v^2}{2} + \frac{1}{2} \left( \frac{P}{2} \right)^2 \right).
\]

The terms with \( v \) on the r.h.s. can be neglected in the limit \( P \to 0 \) and we obtain the condition \( J_S + J_T - 2 > 0 \) which was found in [4] by solving a two-site problem. The above result seems to imply that the combined excitation also sets the lowest energy scale for the elementary excitations. However, we show below that it actually never is the lowest one and in the thermodynamic limit either the SS or the TT (or both) bound states have a lower dispersion in the whole Brillouin zone. The energies of the latter are \( E_{SS} = (J_S + 1)\sin^2(P/2) \) and \( E_{TT} = (J_T + 1)\sin^2(P/2) \) respectively. The long wavelength solution for the ST follows from (17)

\[
E_{ST} = 2(2 + J_S + J_T)\sin^2\left( \frac{P}{4} \right) - \frac{1}{2} \left( \frac{2 + J_S + J_T}{J_S + J_T - 2} \right)^2 \left( \frac{P}{2} \right)^8 + \ldots
\]
Therefore at small momenta at least one of the splittings of the respective dispersions is always positive

\[ E_{ST} - E_{SS} = \frac{1}{2} \left( \frac{P}{2} \right)^2 \left( J_T - J_S \right) + \frac{P^4}{384} \left( 6 + 7J_S - J_T \right) + \ldots \]

\[ E_{ST} - E_{TT} = \frac{1}{2} \left( \frac{P}{2} \right)^2 \left( J_S - J_T \right) + \frac{P^4}{384} \left( 6 + 7J_T - J_S \right) + \ldots \]

These splittings become larger towards the edge of the Brillouin zone.

It was pointed out above that for the combined excitations the differences due to symmetry of the excited state are much less pronounced than for the SS excitations. In one dimensional systems such differences are relatively small and vanish very fast in the limit \( N \to \infty \). For instance, the long wavelength instability of the antisymmetric two-magnon bound state occurs in the region where the so called string hypothesis breaks down (see, e.g., [8],[9]). The origin of this instability can be explained as follows. The flipped spins tend to be located nearby with a probability distribution decaying with lattice distance. The quantum nature of the excitation allows for an "antibonding" (antisymmetric) state. The nodal point of this state corresponds to large separation of flipped spins. Reaching such a separation becomes possible for a long wavelength excitation and therefore leads to the dissociation into scattered states [10]. Thus, the critical region is in fact determined by the vanishing of the dynamic interaction. However, beyond the critical point these states remain special in the sense that they still conserve some solitonic features even within the band of scattered states. For instance, they have the lowest energy and the most flat dispersion or a "heavier mass", which leads to a sharp density of states. In higher dimensions one expects that such excitations would become resonance states in the limit \( N \to \infty \) [11],[12]. The different nature of magnons and orbitons leads to an oscillatory character of the wave function at all momenta. This causes the dynamic interaction to vanish not only at long but also at short wavelengths irrespective of the symmetry. Also for higher dimensional systems only small differences due to symmetry should be expected for an ST excitation. This is in contrast with purely spin excitations, where bound states of different symmetry are well separated in energy [11],[7]. Instead, several closely lying resonances should be observed inside the continuum of scattering states.

4. DISCUSSION

The exact solution for a finite chain presented above has allowed to reveal several distinctive properties of the combined spin-orbiton excitations which are important for the understanding of their nature. It suggests that in a realistic situation these excitations are more likely to be found in a resonant state within the continuum of scattering states, than as true bound states. Despite setting stronger restrictions on the stability of the ordered phase, the combined spin-orbiton excitations have a higher energy and are less stable than excitations of the spin or orbital subsystems. This however opens a possibility to probe the orbiton dynamics via excitations of the spin subsystem, e.g., by neutron scattering. Coupling to the orbital degrees of freedom would produce then resonances in the magnon excitation spectrum when the magnon dispersion crosses with the ST resonance. The overall effect of the band of scattering states on the one magnon dispersion is yet to be studied. But the exact solution for a discrete chain reveals a qualitative difference of the energy spectrum in such bands as compared to the magnon bands. Interaction in magnon bands produces a uniform shift of noninteracting magnons without changing their structure. The ST bands, on the contrary, undergo a radical change from smooth magnon-like in the absence of interaction to a layered structure with ridge shaped individual sub-bands. Such a structure can have a large effect on renormalizing the magnon spectrum, especially at short wavelengths, where oscillations in the wave function and energy dispersion are the largest. The splitting of orbital energy levels due to anisotropy, crystal field etc. would shift the spectrum of ST excitations to higher energies without changing the intrinsic structure of the bands.
ACKNOWLEDGEMENTS. Financial support from the Supreme Council for Scientific and Technological Development of Moldova is acknowledged.

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