Low-Lying Magnetic Excitation of the Shastry-Sutherland Model

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By using perturbation calculation and numerical diagonalization, low-energy spin dynamics of the Shastry-Sutherland model is investigated paying particular attention to the two-particle coherent motion. In addition to spin-singlet- and triplet bound states, we find novel branches of coherent motion of a bound quintet pair, which are usually unstable because of repulsion. Unusual dispersion observed in neutron-scattering measurements are explained by the present theory. The importance of the effects of phonon is also pointed out.

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In recent years, low-dimensional spin systems with a spin gap have been a subject of extensive research. Among them, a two-dimensional antiferromagnet \( \text{SrCu}_2(\text{BO}_3)_2 \) is outstanding in its unique features. These include, (i) spin-gapped behavior \( \langle \cdot \rangle \), (ii) magnetization plateaus \( \langle \cdot \rangle \), and (iii) unusual low-energy dynamics \( \langle \cdot \rangle \).

In Ref. \[4\], it was pointed out that \( \text{SrCu}_2(\text{BO}_3)_2 \) may be modeled by the \( H = 1/2 \) Heisenberg model on the Shastry-Sutherland (SS) lattice \( \langle \cdot \rangle \) (Shastry-Sutherland model, hereafter). This sparked experimental- and theoretical researches on interesting features of the Shastry-Sutherland model \[4\].

Strong geometrical frustration of the SS model allows a simple dimer-product to be the exact ground state \( \langle \cdot \rangle \). In the zeroth-order approximation, a triplet excitation above the dimer-singlet ground state is created by promoting one of the dimer singlets to triplet. In this letter, we consider such particle-like excitations and show that interesting two-particle motion (bound states) is possible because of unusual dynamical properties.

The unit cell of the SS lattice contains two mutually orthogonal dimer bonds (we call them A and B; see Fig.1) and the Hamiltonian can be written as a sum of local Hamiltonians acting only on either A- or B dimers and those acting on both A and B dimers:

\[
\mathcal{H} = \sum_{\alpha=A,B} \mathcal{H}_\alpha + \sum_{\langle \alpha,\beta \rangle=\langle A,B \rangle} \mathcal{H}_{\alpha,\beta}.
\]

In terms of the hardcore triplet bosons \( t \) on dimer bonds \( \langle \cdot \rangle \), the local Hamiltonians are given by \( \mathcal{H}_\alpha = J[-I + t_\alpha \cdot t_\alpha] \) and \( \mathcal{H}_{\alpha,\beta} = \frac{J}{2} \left[ |\text{sign}(\alpha,\beta)| (t_\alpha \cdot t_\beta + (\text{h.c.})) + T_\alpha \cdot T_\beta \right] \), where \( T_\alpha \cdot T_\beta \) denote the \( S = 1 \) operators and the sign factor \( |\text{sign}(\alpha,\beta)| = 1 \) when the arrow on a horizontal bond \( (\beta) \) is emanating from the vertical one \( (\alpha) \) and \(-1 \) otherwise (see Fig.1).

As is easily seen, a unique geometry of the SS lattice allows neither (bare) one-particle hopping \( \langle t^\dagger(x) \cdot t(y) \rangle \) nor pair creation/annihilation of triplets; non-trivial one-particle (triplet) hopping is generated only perturbatively \( \langle \cdot \rangle \) (it occurs at \((J'/J)^6 \) and higher).

**Correlated hopping**

Although one-particle hopping is strongly suppressed, the situation is dramatically different for two-particle cases. The 3-point vertices \( (e.g. \ t^\dagger t \times t) \) contained in the Hamiltonian make non-trivial two-particle hopping like Fig.2 possible already at \((J'/J)^2 \). Note that only one of the two particles hops and that the other is at rest merely to assist the hopping. Therefore, we call such processes **correlated hoppings**; two triplets close to each other can use this new channel of two-particle motion to form various bound states. Although the relevance of correlated hoppings in the SS model was pointed out in \[8\] in the context of magnetization process, the effects of it would be most highlighted in the low-energy dynamics.

Usually, correlated motions are only higher-order corrections to the dominant one-particle processes. However, if the one-particle processes are strongly suppressed for some reasons, correlated ones would play an important role in the low-energy dynamics. The Shastry-Sutherland model is a candidate for such systems. Below we demonstrate that the correlated hoppings can explain the unusual dispersion observed in inelastic neutron-scattering (INS) experiments \[3\].

**Two-triplet motion**

Collecting all the two-particle processes up to \((J'/J)^3 \), we found that the two-particle coherent motion is closed within the four states (Fig. 2) which are decoupled from other states where two triplets are far apart. Because of this special property, if we take the following four relative configurations (Fig. 2)

\[
|\psi(p_x,p_y)\rangle = \langle a(p), b(p), c(p), d(p) \rangle
\]

as the basis, computation of the two-triplet spectra reduces to diagonalization of a four-by-four (up to 3rd order) hopping matrix. Although the form of it is the same as that given in Ref. \[1\], the elements now are functions of spin-1 operators \( T_1 \) and \( T_2 \) of the two triplets; for ex-
ample, the interactions $V_{NN}$ and $V_{NNN}$ between nearest-neighbor- and next-nearest-neighbor pair are given by

$$V_{NN} = \left( \frac{1}{2} J' - \frac{(J')^2}{4J} - \frac{(J')^3}{2J^2} \right) T_1 \cdot T_2$$

$$- \left( \frac{(J')^2}{4J} + \frac{(J')^3}{8J^2} \right) (T_1 \cdot T_2)^2 + \left( \frac{(J')^2}{J} + \frac{(J')^3}{2J^2} \right) V_{NNN} = \frac{(J')^3}{4J^2} T_1 \cdot T_2 .$$

(3)

The meaning of the hopping amplitudes $J_{NN}$ and $J_{3rd}$ can be read off from Fig. 2. Note that $p$ is defined with respect to the chemical unit cell differently from that used in Refs. 33. For completeness, we add another two-particle interaction between a 3rd-neighbor pair $(| \cdots |)$:

$$V_{3rd} = \left( \frac{(J')^2}{2J} + \frac{3(J')^3}{4J^2} \right) T_1 \cdot T_2 ,$$

(4)

which creates immobile (at this order) bound pairs with energy $2V_0 (J', J') + V_{3rd}$ for $S_{tot} = 0, 1$. In the perturbative regime, a pair with total spin $S_{tot} = 0$ or 1 feels attraction whereas one with $S_{tot} = 2$ repels each other, which is the origin of magnetization plateaus of the SS model 33. The occurrence of attraction for a singlet-or triplet pair is not restricted to the SS system and indeed is responsible for the bound states in the 2-leg ladder 13 and the $S = 1/2$ dimerized chain 34. Interchange of the sub(dimer)lattice $A \rightarrow B ((p_x, p_y) \mapsto (p_y, -p_x))$ gives another hopping matrix corresponding to different bound states. Diagonalizing them, we obtain 8 branches (4 for each dimer sublattice) for a given value of total spin. Of course, on top of them, there are dispersionless bound states corresponding to $V_{3rd}$ and infinitely degenerate (up to 5th order) levels corresponding to two isolated triplets at $\omega = 2V_0 (J, J')$. At higher orders of perturbation, the infinite degeneracy will be lifted and this level becomes a narrow continuum. The dispersion curves obtained this way are shown in Fig. 3-5 for $J'/J = 0.5$. Note that the entire spectrum is invariant under $D_{2d}$-operations $(p_x, p_y) \mapsto (p_y, p_x), (p_x, p_y) \mapsto (-p_x, p_y)$ and their products.

As is expected, there appear stable branches of bound states (bold solid lines and dotted lines) below the two-particle threshold (thin broken lines) for $S_{tot} = 0$ and 1. Because of a strong attraction between two triplets on adjacent dimer bonds, the energy decrease from the two-particle threshold is largest for the singlet bound states (Fig. 3). As a direct consequence of the correlated hopping starting from $(J'/J)^2$, the dispersion of the bound states is relatively large compared with that of the lowest single-triplet excitation, which is less than 0.01$J$ for $J'/J = 0.5$. To supplement the perturbation theory (PT) we also performed exact diagonalization (ED) of the original Hamiltonian for finite clusters with 16 and 24 sites under periodic boundary conditions. For small $J'/J$, e.g.

$0.2$, we have confirmed that the results of ED for $S_{tot} = 0$ and 1 are consistent with those of PT.

We investigated excited states for larger $J'/J$ as well. While the bandwidth becomes larger for most branches, an almost flat band was found even for relatively large $J'/J$. For small $J'/J$, the energy obtained by ED is close to that of the 3rd-neighbor bound pairs $(2V_0 + V_{3rd})$ calculated by PT. Therefore we may identify this flat band with the aforementioned 3rd-neighbor bound pairs.

What is more interesting is that although a quintet pair feels repulsion several branches lie slightly below the two-particle threshold. This binding beginning at $(J'/J)^3$ is of purely dynamical origin and is peculiar to the orthogonal dimer systems. Its physical implications to magnetization processes are discussed in detail in Ref. 17. For $S_{tot} = 2$ it is difficult to compare the results of ED with those of PT, because there are so many levels around the two-particle threshold. However, the minimum energy lies below the two-particle threshold as is suggested from PT. We examined numerical data for various $J'/J$ and found that this seems to be the case at least up to $J'/J \approx 0.55$, beyond which no decisive conclusion was drawn due to finite-size effects.

Quite recently, Nojiri et al. found in the ESR (electron spin resonance) spectra a quintet branch at about 1400GHz, which is slightly smaller than twice the single triplet gap 722GHz 10. We believe that our findings are of direct relevance to this observation.

**Selection Rules**

The unique structure of the SS lattice allows only a few branches to be observed in INS 3. The Fourier transform $S^j(p) \ (j = x, y, z)$ of local spin operators create single-dimer-triplet states

$$S^j(p)|G.S.\rangle = f_+(p)|j; A(p)\rangle + f_-(p)e^{-i\frac{1}{2}(p_x+p_y)}|j; B(p)\rangle$$

(5)

over the dimer ground state, where $|j; A/B(p)\rangle \equiv t^b_{A/B,j}(p)|G.S.\rangle$. Since one of the structure factors $f_{\pm}(p) = \mp isin(l_d(p_x \pm p_y)/2\sqrt{2})$ vanishes on $p_x = \mp p_y$, only an A (B) triplet is excited along the line $p_x = p_y (p_x = -p_y)$. Although the state $S^j(p)|G.S.\rangle$ contains only a single triplet, the first-order perturbation (3-point vertices) broadens the wave function and $S^j(p)|G.S.\rangle$ can have a finite overlap with the two-triplet states. Since $S^j(p_x, \pm p_y)|G.S.\rangle$ is even under the reflection $\sigma_d(1, \pm)$ (i.e. belongs to $\Sigma_1$-representation), any states which are connected to $S^j(p_x, \pm p_y)|G.S.\rangle$ by perturbation should also be even. From this, it follows that any bound-state wave functions which contain “b” and “c” in an antisymmetric manner (i.e. $\Sigma_2$ are orthogonal to $S^j(p)|G.S.\rangle$; that is, INS experiments performed along the [110] ([110]) direction (as in Ref. 3) observe only bound states shown by solid black lines in Fig. 4. Qualitative features agree with what was observed in experiments 3.
To compare our results with experiments quantitatively, we carried out ED since the value \( J'/J = 0.635 \) recently estimated \[15\] for \( \text{SrCu}_2(\text{BO}_3)_2 \) is too large for the results of PT to be used. The results for \( J = 85 \text{K} \) are shown in Fig. 6. By close inspection of the wave function obtained both by PT and ED, we can identify the INS-active branches shown by the lower black lines in Fig. 4 in the numerical spectrum for \( J'/J = 0.635 \). The energies thus obtained are plotted by open circles in Fig. 6 giving good agreement with the experimental value (filled circles) \[15\]. In particular, the energy of the lowest INS-active two-triplet is given by 4.99\text{meV} at \( \mathbf{p}=0 \). This agrees with 5.0meV observed in experiments \[2\] and supports the validity of the value \( J'/J = 0.635 \) for \( \text{SrCu}_2(\text{BO}_3)_2 \).

This selection rule does not exclude the possibility of observing the remaining branches by other methods. For example, optical methods probe excitations at \( \mathbf{p} = 0 \). The representations (\( E, B_2 \)) and (\( A_1, A_2, B_1, B_2, E \)) are active in far infrared spectroscopy (FIR) \[15\] and Raman scattering \[14\], respectively. Analysis of the wave functions shows that, for example, the 1-triplet, the lowest 2-triplet (singlet pair), and the second-lowest 2-triplet (3rd-neighbor singlet pair) in Fig. 3, 4 belong to \( E, A_2 \oplus B_1, A_1 \oplus B_2 \), respectively. This is qualitatively consistent with the results of the experiments. Moreover, analytical and numerical results suggest that the lowest bound state in the triplet sector is dominated by the 3rd-neighbor pair (E-rep), which is not excited by \( S^2(\mathbf{p}) \); using the above parameters, the energy of it (at \( \mathbf{p}=0 \)) is given by 36.5cm\(^{-1} \), which does not contradict the FIR- and ESR results. Detailed analyses will be published elsewhere.

**Phonons**

Up to now, we have treated dynamics of a purely magnetic system. In real materials, coupling between phonons and spin degrees of freedom would be important. To see how inclusion of virtual phonons yields effective interactions, we treat only the simplest case of the Einstein phonon where each dimer (\( J \)) bond rotates independently around its equilibrium position with a frequency \( \omega_0 \). In contrast to the purely magnetic case, pair creation/annihilation and one-particle hopping which accompany emission of a phonon are allowed.

Here we only consider the most important case of a nearest-neighbor pair. A little algebra shows that the second-order processes generate an attractive interaction \( V_{NN}^{(\text{ph})} \propto -(2J + \omega_0)/(J + \omega_0) \) even for a quintet pair; a singlet pair feels attraction while a triplet repulsion. Therefore, we may expect that the lowering in energy for quintet bound states is enhanced by phonons.

In \( \text{SrCu}_2(\text{BO}_3)_2 \) the interlayer coupling \( J_L \) is not negligible either. However, \( J_L \) changes nothing as far as \textit{intra}-layer excitations are considered \[15\]. It might make a new (INS-inactive) interlayer bound states slightly below the two-particle threshold by a very weak interaction \( J_L(1 - 1/4(J'/J)^2) \mathbf{T}_1 \cdot \mathbf{T}_2 \). These bound states have a small bandwidth of the order \( (J'/J)^6 \).

In conclusion, we demonstrated that unique dynamical property of the SS model allows an interesting pairing of triplets. Bound two triplets can move on a lattice much easier than a single isolated triplet can and this fact explains the recent INS experiments. Note that the difference in dispersion is not quantitative and originates from that in hopping mechanisms. Moreover, it enables the two triplets to form quintet ‘bound’ states despite the repulsion between them. To complement the perturbative consideration, we also carried out exact diagonalization and obtained for \( J'/J = 0.635 \) the results consistent with those of experiments.

After this work was completed, we became aware of a preprint by Knetter et al. \[cond-mat 0005322\]. They investigated gaps at \( \mathbf{p}=0 \) for singlet- and triplet bound states using a similar but different method.

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CAPTIONS

Fig. 1
Configurations of two orthogonal dimers in a unit cell. If the direction of the vertical (α) bond is fixed (say, pointing downward), two configurations (left and right) are allowed for the horizontal (β) bond.

Fig. 2
“Hopping” processes of two triplets. A(B) dimers are shown by bold black (gray) lines. A filled circle denotes a representative point of a unit cell r. Symbols for the matrix elements are the same as those in Ref. [17].

Fig. 3
Singlet (S_{tot} = 0) dispersion in the [110]- and [100]-direction for J'/J = 0.5 obtained by the perturbation. Dashed- and dotted lines denote B-branches and 3rd-neighbor bound states, respectively. Two-particle threshold lies at 1.356J (thin broken line).

Fig. 4
Same for triplet (S_{tot} = 1) sector. Only branches shown by solid black lines are observable in neutron-scattering experiments.

Fig. 5
Same for quintet (S_{tot} = 2) sector. Note that several branches lie below 2-particle threshold (thin broken line). Because of higher-order processes, states above the 2-particle threshold (thin dashed line) are unstable.

Fig. 6
The branches with S_{tot} = 1. The results of the ED for J'/J = 0.635 and J = 85 K are shown by open symbols. Among them the branches observable in neutron-scattering experiments are shown by the open circles (lower: 1-triplet, upper: 2-triplet). For comparison, the experimental results are shown by the closed circles. Small splittings at p=0 are artifacts coming from the shape of a cluster (which is C_2-invariant) used in ED.
Fig. 1 Totsuka, Miyahara, and Ueda

Fig. 2 Totsuka, Miyahara, and Ueda
\[ \left( \pi, \pi \right) \]
Fig. 4 Totsuka, Miyahara, and Ueda
Fig. 5 Totsuka, Miyahara, and Ueda
Fig. 6 Totsuka, Miyahara, and Ueda