Dispersion and Symmetry of Bound States in the Shastry-Sutherland Model

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Bound states made from two triplet excitations on the Shastry-Sutherland lattice are investigated. Based on the perturbative unitary transformation by flow equations quantitative properties like dispersions and qualitative properties like symmetries are determined. The high order results [up to $(J_2/J_1)^{14}$] permit one to fix the parameters of $SrCu_2(BO_3)_2$ precisely: $J_1 = 6.16(10)$ meV, $x := J_2/J_1 = 0.603(3)$, $J_\perp = 1.3(2)$ meV. At the border of the magnetic Brillouin zone a general double degeneracy is derived. An unexpected instability in the triplet channel at $x = 0.63$ indicates a transition towards another phase. The possible nature of this phase is discussed.

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Quantum antiferromagnets are at the center of research not only because of the high T_c superconductors. Of particular interest are systems which do not have an ordered, Néel-type ground state. Their ground state is a spin liquid without long range spin order. Spin liquids are favored by low spin ($S = \frac{1}{2}$ mostly), low coordination number ($Z \in \{2,3,4\} \Rightarrow D \in \{1,2\}$), and strong geometric frustration.

Dimer solids are transparent cases of spin liquids. In $D = 1$, the generic example is the Majumdar-Ghosh model [1] of which Shastry and Sutherland found a $D = 2$ generalization (ShaSu model) [2]. In both cases frustration is essential. Each spin is coupled to pairs of spins (dimers). If these pairs form singlets, the couplings between dimers is without effect and the singlet-on-dimers product state is always an eigenstate and for certain parameters the ground state [2–5]. The systems are gapped. The elementary excited states are dressed $S = \frac{1}{2}$ ($D = 1$) [4] or $S = 1$ ($D = 2$) entities. They interact strongly and form bound and antibound states in various spin channels.

Because of its recent realization in $SrCu_2(BO_3)_2$ [6,7] the ShaSu model (Fig. 1) is presently attracting enormous interest. The Hamiltonian reads

$$H(J_1, J_2) = J_1 \sum_{\langle \mathbf{i},\mathbf{j}\rangle \text{dimer}} \mathbf{S_i S_j} + J_2 \sum_{\langle \mathbf{i},\mathbf{j}\rangle \text{square}} \mathbf{S_i S_j}\,. \quad (1)$$

In this Letter we start from the dimer phase [5]. We focus on bound states formed from pairs of the elementary triplets and their symmetries, degeneracies, and dispersion. The perturbative unitary transformation [8] based on flow equations [9] enables us to link smoothly and uniquely $H(J_1, J_2)$ at $x := J_2/J_1 \neq 0$ to an effective H_{eff} conserving the number of triplets on dimers $[H_{\text{eff}}, H(J_1, 0)] = 0$. This permits a clear distinction between the ground state (without triplets), the 1-triplet sector, the 2-triplet sector, etc.

In terms of H_{eff} the dynamics of one triplet is hopping $t_{h;\mathbf{i}}$ ($t_{v;\mathbf{i}}$) starting from a horizontal (vertical) dimer by i_x dimers right and i_y dimers up. Nothing else is possible due to triplet number conservation. The elements t are computed in order 15 [5,10,11].

The dynamics of two triplets at large distances is governed by 1-triplet hopping. At smaller distances a two-particle interaction occurs additionally given by $W_{h;\mathbf{d};\mathbf{i},\mathbf{d}'}$ ($W_{v;\mathbf{d};\mathbf{i},\mathbf{d}'}$) starting with one triplet on a horizontal (vertical) dimer and another at distance $\mathbf{d}$. The action of H_{eff} is to shift the triplets to $\mathbf{i}$ and to $\mathbf{i} + \mathbf{d}'$. Nothing else is possible due to triplet number conservation. Since the total spin is conserved ($S \in \{0,1,2\}$) the distances are restricted to $\mathbf{d}, \mathbf{d}' > \mathbf{0}$, i.e., $d_x > 0$ or $d_x = 0 \wedge d_y > 0$, because the exchange parity is fixed.

The action of H_{eff} yields the *combined* effect of hopping and interaction denoted by $A_{\mathbf{d};\mathbf{i},\mathbf{d}'}$. The true 2-triplet interaction is easily found by subtracting the 1-triplet hopping [5,10,11]

$$W_{\mathbf{d};\mathbf{0},\mathbf{d}'} = A_{\mathbf{d};\mathbf{0},\mathbf{d}'} - t_{\mathbf{d}'-\mathbf{d}} - \delta_{\mathbf{d}',\mathbf{d}} t_{\mathbf{0}}\,, \quad (2a)$$

$$W_{\mathbf{d};\mathbf{d}-\mathbf{d}',\mathbf{d}'} = A_{\mathbf{d};\mathbf{d}-\mathbf{d}',\mathbf{d}'} - t_{\mathbf{d}-\mathbf{d}'} - \delta_{\mathbf{d}',\mathbf{d}} t_{\mathbf{0}}\,, \quad (2b)$$

$$W_{\mathbf{d};-\mathbf{d}',\mathbf{d}'} = A_{\mathbf{d};-\mathbf{d}',\mathbf{d}'} - t_{-\mathbf{d}-\mathbf{d}'}\,, \quad (2c)$$

$$W_{\mathbf{d};\mathbf{d},\mathbf{d}'} = A_{\mathbf{d};\mathbf{d},\mathbf{d}'} - t_{\mathbf{d}+\mathbf{d}'} \quad (2d)$$

(distinction h/v omitted for clarity). Otherwise A and W are identical. The coefficients W for $S \in \{0,1,2\}$ yield

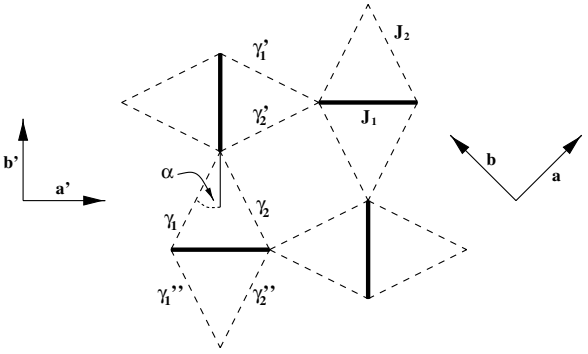

FIG. 1. Unit cell of the Shastry-Sutherland (ShaSu) lattice as realized in $SrCu_2(BO_3)_2$, where $(\mathbf{a}, \mathbf{b})$ are unit vectors; $(\mathbf{a}', \mathbf{b}')$ and the coefficients γ are used to analyze Raman scattering.

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the complete two-particle dynamics. We compute W up to x^{12} and for the lowest-lying states even up to x^{14}.

During the virtual processes [12] the triplet number is changed. Because of the frustration of the ShaSu lattice H in (1) changes the number of triplets on the dimers at most by one [7,10,11]. An excitation or a deexcitation on a horizontal (vertical) dimer is possible iff at least one of the vertical (horizontal) dimers on the left and right (above and below) are excited. This restriction implies that one triplet hops only in x^6 (cf. figures in [11,13]).

The motion of two triplets together is much less restricted (cf. Fig. 2). Matrix elements occur in x^2 as first observed for total spin $S = 2$ [14]. But the dispersion of bound states starts only in x^3 (contrary to x^4 claimed in Ref. [15]). Two adjacent triplets interact linearly in x ($-x$ for $S = 0$, $-x/2$ for $S = 1$, $x/2$ for $S = 2$). The energy of the initial and final states in each row in Fig. 2 differs by $\mathcal{O}(x)$. Hence both rows must be combined making it an $(x^2)^2/x = x^3$ process eventually. This applies to the 8 (anti)bound states derived from two triplets on (next) nearest neighbor dimers. The dispersion of any other state sets in at higher order.

We use the following basis for the 2-triplet states:

$$|\mathbf{k}, \mathbf{d}, \sigma\rangle := N^{-1/2} \sum_{\mathbf{r}} e^{[i(\mathbf{k}+\sigma\mathbf{Q})(\mathbf{r}+\mathbf{d}/2)]} |\mathbf{r}, \mathbf{r} + \mathbf{d}\rangle , \tag{3}$$

where $\mathbf{k}$ is the conserved total momentum in the magnetic Brillouin zone (MBZ) applying due to the two sublattices; $\sigma \in \{0, 1\}$, $\mathbf{Q} := (\pi, \pi)$, N is the number of dimers, $|\mathbf{r}, \mathbf{r} + \mathbf{d}\rangle$ denotes the state with triplets at $\mathbf{r}$ and at $\mathbf{r} + \mathbf{d}$. The distance $\mathbf{d}$ is restricted $\mathbf{d} > \mathbf{0}$, i.e., $d_x > 0$ or $d_x = 0 \wedge d_y > 0$. The matrix elements of H_{eff} in the basis (3) are real due to translation and inversion symmetry.

Before the quantitative analysis of the bound states a qualitative aspect, a general double degeneracy at the border of the MBZ, shall be derived. To see this, consider the combination of a shift by the dimer-dimer spacing along $\mathbf{a}'$ (S), a reflection about $\mathbf{a}$ (R), and the inversion $\mathbf{r} \to -\mathbf{r}$ (I) (cf. Fig. 1). The combinations SR and I are symmetries of the Hamiltonian. For $k_x + k_y = \pi$ (part of the MBZ border) definition (3) implies for the total combination SRI the mapping

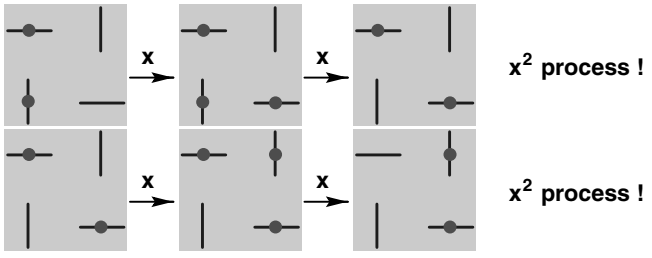

FIG. 2. Leading processes of correlated 2-triplet hopping. Dark dots are triplets; bars are dimers.

$$|\mathbf{k}, \mathbf{d}, \sigma\rangle \to e^{ik_x + i\pi(d_x + d_y + \sigma + PS)} |\mathbf{k}, -\begin{pmatrix} d_y \\ d_x \end{pmatrix}, 1 - \sigma\rangle , \tag{4}$$

where $P \in \{0, 1\}$ being unity iff $-(d_y, d_x) < \mathbf{0}$ so that the triplets must be swapped to pass from $-(d_y, d_x)$ to (d_y, d_x). It is crucial that SRI links $|\mathbf{k}, (d_x, d_y), 0\rangle$ and $|\mathbf{k}, -(d_y, d_x), 1\rangle$ like a 2D rotation $\left(\begin{smallmatrix} 0 & -1 \\ 1 & 0 \end{smallmatrix}\right)$ up to a prefactor. Hence its eigenvectors are complex with linearly independent real and imaginary parts and so are the simultaneous eigenvectors of SRI and H_{eff}. Because H_{eff} is real, the real and the imaginary parts constitute, in fact, linearly independent eigenvectors to the same eigenvalue. The same double degeneracy is concluded for the other parts of the MBZ border by S and 90° rotation (D). It is also valid in the 1-triplet sector [11].

The double degeneracy at the MBZ border is interesting for analyzing experiment, too. Degeneracy reduces the large number of energetically close states helping to resolve different bound states.

Since 1-triplet hopping is of higher order than interaction, an analytic expansion for the energies of the bound states is possible. At finite order in x only configurations contribute where the two triplets are not too far away from each other. Of course, higher orders imply larger, but still finite distances. In particular, the energies of the four states which evolve from neighboring triplets can be computed very well since their interaction is linear. Investigating the matrix elements shows that it is sufficient to study the distances $\mathbf{d} \in \{(0, 1), (1, 0), (1, \pm 1)\}$ for order 5. To x^{14} only $\mathbf{d} \in \{(1, \pm 2), (2, \pm 1), (0, 2), (2, 0), (2, \pm 2)\}$ must be added. So, for given total momentum only a finite 8×8 or 24×24 matrix has to be analyzed. For illustration consider the elements $A_{(0,1);\mathbf{i},(2,1)}$ (the Fourier transform of $\mathbf{i}$ yields the momentum dependence) connecting $(0, 1)$ and $(2, 1)$ which is $\mathcal{O}(x^4)$. By 2nd order perturbation one sees that the resulting energy shift is $(x^4)^2/x = x^7$ only.

The elements connecting shorter distances to longer distances and the elements among longer distances do not need to be known to very high order. Consider again the process $(0, 1) \leftrightarrow (2, 1)$. In order x^7 the element $A_{(0,1);\mathbf{i},(2,1)}$ must be known only in x^4 and $A_{(2,1);\mathbf{i},(2,1)}$ only in x^1; in order x^9 the element $A_{(0,1);\mathbf{i},(2,1)}$ must be known only up to x^6 and $A_{(2,1);\mathbf{i},(2,1)}$ only in x^3, etc.

We have analyzed the dispersions in x^5 of the four states bound linearly in x in the MBZ. Fukumoto's results are mostly confirmed [16,17]. At particular points of high point group symmetry [$(0, 0)$; $(0, \pi)$] the Hamiltonian splits into six blocks corresponding to different representations of the square point group $4mm$. At these points the analysis up to x^{14} is carried out [17]. The symmetries are classified according to the irreducible representations (four 1D, one 2D) of the point group $4mm$ $\Gamma_1(1)$, $\Gamma_2(x^2 - y^2)$, $\Gamma_3(xy)$, $\Gamma_4(xy(x^2 - y^2))$, $\Gamma_5(x, y)$ where simple polynomials are given in brackets to show the transformation behavior.

The extrapolated energies are depicted in Figs. 3 ($S = 0$) and 4 ($S = 1$) as functions of x. For those energies which stay separated from the two-particle continuum Dlog-Padé approximants are used successfully [18]. The results are stable under changes of the polynomial degrees. The energies close to the continuum (here simply twice the gap Δ between the ground state and the elementary triplet at $\mathbf{k} = \mathbf{0}$) are given with less reliability by the truncated series or by a nondefective Dlog-Padé approximant.

In Figs. 3 and 4 the modes are sorted in energetically ascending order for small values of x: solid, long-dashed, and short-dashed curves. The Γ_5 modes are naturally degenerate. The double degeneracy for $\mathbf{k} = (0, \pi)$ does *not* result from the point group but originates from the complex conjugation as explained above. The dash-dotted curve at $(0, \pi)$ has to be compared to the solid and the long-dashed curve to assess the dispersion of these two modes from $\mathbf{0}$ to $(0, \pi)$. While for $S = 0$ this dispersion always has the expected behavior with $\omega(\mathbf{0}) < \omega((0, \pi))$ the energies for $S = 1$ are reversed for small values of x (cf. [16]). Only above $x \approx 0.55$ the relation $\omega(\mathbf{0}) < \omega((0, \pi))$ holds for $S = 1$.

We do not agree with Ref. [16] that the two lowest states are of s-wave–type since this would imply that they are Γ_1. Instead, the $S = 0, 1$ states are odd under reflection about $\mathbf{a}'$ (R1) or about $\mathbf{b}'$ (R2) (cf. Fig. 1). For $S = 0$, the lowest state is even under SD and the second lowest is odd. For $S = 1$, it is vice versa. The Γ_5 states can be viewed as being of p-wave–type.

For $S = 0$, the lowest mode vanishes at the same x as does the gap Δ. No additional instability occurs for $S = 0$. There is, however, a salient instability for $S = 1$ (Fig. 4) at $x = 0.63$. This comes as a surprise since one expects binding effects in antiferromagnets to be largest for $S = 0$. The singularity at $x = 0.630(5)$ is very stable occurring in all nondefective Dlog-Padé approximants. The softening of the $S = 1, \Gamma_3$ mode at $x = 0.63$ in Fig. 4 implies a level crossing with the single triplet at $x = 0.62$. The latter, however, is Γ_5 at $\mathbf{k} = \mathbf{0}$ [11] so that no level repulsion prevents the crossing [21].

A mode softening like the one at $x = 0.63$ might indicate a 2nd order transition. But we find the softening of a *bound* mode of two triplets proving significant attraction. If this attraction extends also to three and more triplets binding them to any total spin, the transition would be 1st order towards a condensate of triplets occurring *below* 0.63. Interestingly, by Schwinger boson mean-field approximation a 1st order transition towards a helical phase was found at $x \approx 0.61$ [22]. But the nature of the new phase is as yet unsettled. A 1st order transition at $x = 0.677$ into a gapped singlet phase extending up to a 2nd order transition at $x = 0.86$ was proposed [23]. But our instability occurs at a much lower value of x and the antiferromagnetic magnetization at $x = 0.86$ is significantly finite [10] in contradiction to a *second* order phase transition to a phase without long-range order (as mentioned already in Ref. [23]). So the transition order and the intermediate phase still deserve further investigation.

Next we determine x and J_1 for $SrCu_2(BO_3)_2$. The steep decrease of the bound $S = 1$ state enables us to fix x very precisely. Based on ESR [20], FIR [24], as well as INS [19], we assume $\Delta = 2.98$ meV and $\omega|_{S=1} = 4.7$ meV leading to $x = 0.603(3)$ and $J_1 = 6.16(10)$ meV. The 1-triplet dispersion is in excellent agreement with experiment (cf. inset of Fig. 3 and Ref. [11]). Raman scattering [15] provides further strong support because the energy of the Γ_3 singlet matches 30 cm^{-1} perfectly. The Γ_4 singlet at 25 cm^{-1} [25] is forbidden by symmetry since the Raman operator is effectively Γ_3.

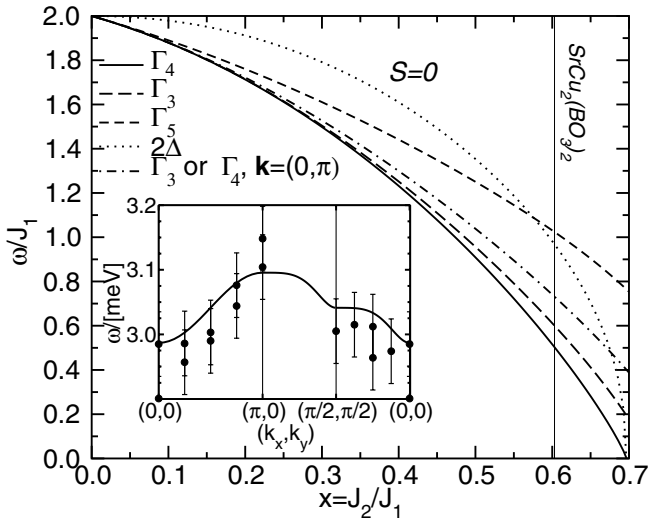

FIG. 3. Energy of the lowest-lying $S = 0$ states. Curves refer to $\mathbf{k} = \mathbf{0}$ except the dash-dotted one. The dotted curve displays the continuum at 2Δ. Inset: 1-triplet dispersion. Theory at $x = 0.603, J_1 = 6.16$ meV; data from Ref. [19], experimental errors at least as large as indicated.

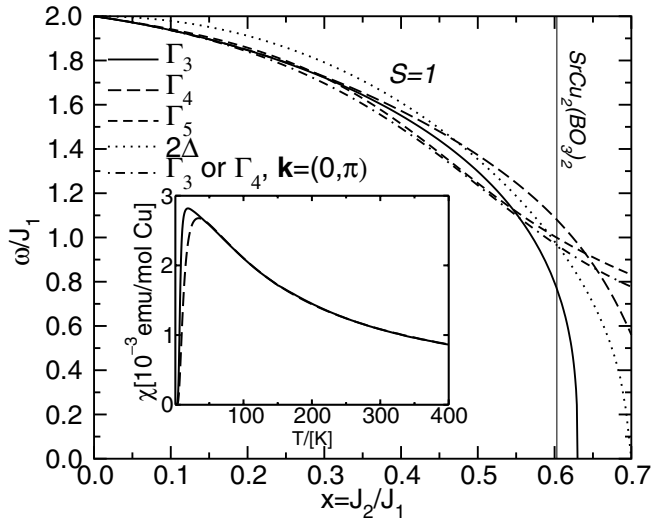

FIG. 4. As in Fig. 3 for $S = 1$. Inset: Magnetic susceptibility. Theory (dashed) with directional rms average $g = 2.13$ [20], x, J_1 as in Fig. 3; experiment (solid) on powder [6].

In leading order t/U the Raman operator $R = \sum \gamma_{\mathbf{i},\mathbf{j}} \mathbf{S_i S_j}$ couples the same spins as the Hamiltonian. But only the antisymmetric ($\gamma_1 = -\gamma_2$) part of R on the dashed bonds (cf. Fig. 1) creates excitations from the ground state. By geometry we have $\gamma_1'' = \gamma_2$ and $\gamma_2'' = \gamma_1$ so that the effective R_{eff} is odd under $R1$ and $R2$. But the projection of the vector potential $\mathbf{A}$ ($\mathbf{A}||\mathbf{E}$) on the bonds under study in the polarization ($\mathbf{ab}$) implies $\gamma_1 = \gamma_2 = -\gamma_1' = -\gamma_2' = \gamma\cos(2\alpha)$ (γ microscopic constant, α angle in Fig. 1), i.e., an even component implying $R_{\text{eff}} = 0$. On the contrary, polarization ($\mathbf{a'b'}$) yields $\gamma_1 = -\gamma_2 = -\gamma_1' = \gamma_2' = \gamma\sin(2\alpha)$ so that $R_{\text{eff}} \neq 0$. This finding agrees nicely with experiment where on $T \to 0$ the intensities almost vanish for ($\mathbf{ab}$) but grow for ($\mathbf{a'b'}$) [15]. Additionally, $\gamma_1 = -\gamma_1'$ and $\gamma_2 = -\gamma_2'$ imply odd parity under SD so that R_{eff} is indeed Γ_3, not Γ_4. Calculating the next $\Gamma_3, S = 0$ bound state (less systematically) yields 45 cm^{-1} in good agreement with the experimental 46 cm^{-1} line, too.

We conclude that the 2D model (1) explains the low-lying excitations of $SrCu_2(BO_3)_2$ perfectly. Thermodynamic quantities like the susceptibility $\chi(T)$ require the inclusion of the interplain coupling $J_\perp$ which is fully frustrated not changing the dimer spins [26]. We have employed a Dlog-Padé approximant for the $1/T$ expansion of the 2D χ_{2D} [10] complemented by the condition $\Delta = 2.98$ meV. This ansatz works fine for $T > 35$ K. The 3D χ_{3D} is computed from χ_{2D} on the mean-field level as $\chi_{3D}^{-1} = \chi_{2D}^{-1} + 4J_\perp$. The inset of Fig. 4 shows that theory [$J_\perp = 1.3(2)$ meV] and experiment [6] agree without flaw above 40 K. Our value for $J_\perp$ is significantly higher than the one in Ref. [26] due to different values of x and J_1.

The above comprehensive analysis of bound states is a fine example of the efficiency of perturbation by unitary transformation [8] based on flow equations. This clear concept allows one to distinguish uniquely sectors with different particle numbers and other different quantum numbers like the total spin. Here the concept was put to use to analyze the Shastry-Sutherland lattice as realized in $SrCu_2(BO_3)_2$. To our knowledge it is the first quantitative description of two-particle bound states in 2D.

The symmetries of experimentally relevant states were determined. The reliability of the high order results allows one to fix the experimental coupling constants very precisely [$J_1 = 6.16(10)$ meV, $J_2/J_1 = 0.603(3)$, $J_\perp = 1.3(2)$ meV]. Thereby, different experiments [electron spin resonance, far-infrared spectroscopy, inelastic neutron scattering, Raman, $\chi(T)$] are explained consistently. We suggest to exploit the double degeneracy derived here to resolve different bound states at the border of the MBZ.

An unexpected instability for the $S = 1$ 2-triplet bound state is found at $x \approx 0.63$ indicating a transition to another phase at much lower values of x than found in Refs. [5,10,23]. The vicinity of $SrCu_2(BO_3)_2$ to this transition suggests to attempt a closer experimental analysis. Pressure and/or substitution will certainly influence the ratio J_2/J_1. Thereby one may hope to scan through the transition and to examine the unknown phase beyond.

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