Dedicated to Prof. F. Wegner on occasion of his 60th birthday.

Quantum antiferromagnets are at the center of research not only because of the high \( T_c \) superconductors. Of particular interest 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 \( (Z \in \{2, 3, 4\}) \). 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 is \( |\uparrow \uparrow \rangle \). The systems are gapped. The elementary excited states are dressed \( S = \frac{1}{2} \) \((D = 1)\), \( S = 1 \) \((D = 2)\) entities. They interact strongly and form bound and antibound states in various spin channels. Due to its recent realization in \( SrCu_2(BO_3)_2 \), the ShaSu model (Fig. 1) is presently attracting enormous interest. The Hamiltonian reads

\[
H(J_1, J_2) = J_1 \sum_{(i,j)\text{dimer}} S_i S_j + J_2 \sum_{(i,j)\text{square}} S_i S_j .
\] (1)

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

In terms of \( H_{\text{eff}} \) the dynamics of one triplet is hopping \( t_{h,i} \) \((t_{v,i})\) starting from a horizontal \((v)\) 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 \( \{010\} \).

The dynamics of two triplets at large distances is governed by 1-triplet hopping. At smaller distances a 2-particle interaction occurs additionally given by \( W_{h,h,i,d'}\) \((W_{v,v,i,d'})\) starting with one triplet on a horizontal \((v)\) dimer and another at distance \( d \). The action of \( H_{\text{eff}} \) is to shift the triplets to \( i \) and to \( i + d' \). 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 \( d, d' > 0 \), i.e. \( d_x > 0 \) or \( d_x = 0 \wedge d_y > 0 \), because the exchange parity is fixed.

The action of \( H_{\text{eff}} \) yields the combined effect of hopping and interaction denoted by \( A_{d,1,d'} \). The true 2-triplet interaction is easily found by subtracting the 1-triplet hopping \( \uparrow \uparrow \uparrow \uparrow \).

\[
\begin{align}
W_{d,0,d'} &= A_{d,0,d'} - t_{d,d'} - \delta_{d,d'} t_{0} \\
W_{d,d,-d',d'} &= A_{d,d,d'-d'} - t_{d-d'} - \delta_{d,d} t_{0} \\
W_{d,-d',d'} &= A_{d,-d',d'} - t_{d-d'} \\
W_{d,d,d'} &= A_{d,d,d'} - t_{d+d'}
\end{align}
\] (2a)

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

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**Fig. 1.** Unit cell of the Shastry-Sutherland (ShaSu) lattice as realized in \( SrCu_2(BO_3)_2 \). \((a,b)\) are unit vectors; \((a',b')\) and the coefficients \( \gamma \) are used to analyse Raman scattering.
the complete 2-particle dynamics. We compute \( W \) up to \( x^{12} \), the coefficients for the lowest-lying states even up to \( x^{14} \).

During the virtual processes \([12]\) the triplet number is changed. Due to the frustration of the ShaSu lattice \( H \) in \([4]\) changes the number of triplets on the dimers at most by one \([7,10,11]\). An excitation or a de-excitation 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. Figs. in \([3,1]\)).

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. \([11]\)). 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 state in each row in Fig. \([2]\) differ by \( O(x) \). Hence both rows must be combined making it an \( (x^2)^2/x = x^4 \) 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.

\[
|k, \mathbf{d}, \sigma\rangle \rightarrow e^{ik_x+\pi(d_x+d_y+\sigma+PS)}|k, -(d_y, 1-\sigma)\rangle, \quad (4)
\]

where \( P \in \{0,1\} \) being unity iff \( -(d_y, d_x) < 0 \) 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 \([k, (d_x, d_y), 0)\) and \([k, -(d_y, d_x), 1)\) like a 2D rotation \[
\begin{pmatrix}
0 & -1 \\
1 & 0
\end{pmatrix}
\]
up to a prefactor. Hence its eigen vectors are complex with linearly independent real and imaginary part and so are the simultaneous eigen vectors of SRI and \( H_{\text{eff}} \). Because \( H_{\text{eff}} \) is real the real and the imaginary part constitute in fact linearly independent eigen vectors to the same eigen value. The same double degeneracy is concluded for the other parts of the MBZ border by \( S \) and \( 90^\circ \) rotation \((D)\). It is also valid in the 1-triplet sector \([1]\).

The double degeneracy at the MBZ border is interesting for analysing 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 \( \times \) 24 \( = \) 192 matrix has to be analysed. For illustration consider the elements \( A_{(0,1),(2,1)} \) (the Fourier transform of \( i \) yields the momentum dependence.) connecting \((0,1)\) and \((2,1)\) which is \( O(x^4) \). By second order perturbation one sees that the resulting energy shift is \( (x^2)^2/x = x^7 \) only.

Furthermore, 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 \( A_{(0,1),(2,1)} \) must be known only in \( x^4 \) and \( A_{(2,1),(2,1)} \) only in \( x^4 \); in order \( x^9 \) the element \( A_{(0,1),(2,1)} \) must be known only up to \( x^6 \) and \( A_{(2,1),(2,1)} \) only in \( x^3 \) and so on.

We have analysed the dispersions in \( x^5 \) of the four states bound linearly in \( x \) in the MBZ. Fukumoto’s results are mostly confirmed \([4,7]\). At particular points of high point group symmetry \((0,0),(0,\pi),(\pi/2,\pi/2)\) 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 \([7]\). 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, 4 (S = 0) and 5 (S = 1) as functions of x. For those energies which stay separated from the 2-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 $\omega < \omega_0$) the energies for $S = 0$ are given with less reliability by the truncated series or by a non-defective Dlog-Padé approximant.

![Image](image_url)

**FIG. 3.** Energy of the lowest-lying $S = 0$ states. Curves refer to $k = 0$ except the dashed-dotted one. The dotted curve displays the continuum at $2\Delta$. Inset: 1-triplet dispersion. Theory at $x = 0.603, J_1 = 6.16$ meV; data from Ref. [18], experimental errors at least as large as indicated.

In Figs. 3, 4 the modes are sorted in energetically ascending order for small values of $x$: solid, long dashed, and short dashed curves. The $\Gamma_5$ modes are naturally degenerate. The double degeneracy for $k = (0, \pi)$ does not result from the point group but originates from the complex conjugation as explained above. The dashed-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 0 to $(0, \pi)$. While for $S = 0$ this dispersion always has the expected behavior with $\omega(0) < \omega((0, \pi))$ the energies for $S = 1$ are reversed for small values of $x$ (cf. Fig. 4). Only above $x \approx 0.55$ the relation $\omega(0) < \omega((0, \pi))$ holds for $S = 1$.

We do not agree with Ref. [10] that the two lowest states are of $s$-wave type since this would imply that they are $\Gamma_1$. Instead the $S = 0, 1$ states are odd under reflection about $a^* (R1)$ or about $b^* (R2)$ (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 $\Gamma_5$ 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 $\Delta$. So no additional instability occurs for $S = 0$. This provides evidence against a competing singlet phase as presumed in Ref. [21]. There is, however, a salient instability for $S = 1$ (Fig. 3) at $x = 0.63$. This comes as a surprise since one expects in antiferromagnets binding effects to be largest for $S = 0$. The singularity at $x = 0.630(5)$ is very stable occurring in all non-defective Dlog-Padé approximants. We take the vanishing of a bound 2-triplet state with $S = 1$ as evidence for strong attraction between triplets which are neither parallel ($S = 2$) nor antiparallel ($S = 0$). The attraction points towards a first order transition into a condensate of triplets occurring at much lower $x$ than thought previously [1]. The angle being neither zero nor $\pi$ corroborates a helical order if one adopts a classical view [2, 3]. In the light of the instability for $S = 1$ we interpret the findings by Koga and Kawakami [21] as indication of the same instability. It is so far not excluded that the bulk triplet condensate (the helical phase) is a singlet or that it can be linked to a singlet [2, 3].

Next we determine $x$ and $J_1$ for SrCu$_2$(BO$_4$)$_2$. 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 in Fig. 3 and Ref. [1]). Raman scattering [15] provides further strong support because the energy of the $\Gamma_3$ singlet matches 30 cm$^{-1}$ perfectly. The $\Gamma_4$ singlet at 25 cm$^{-1}$ [25] is forbidden by symmetry.
since the Raman operator is effectively $\Gamma_3$.

In leading order $t/U$ the Raman operator $R = \sum \gamma_{ij} \mathbf{S}_i \mathbf{S}_j$ 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 $\gamma_1^\prime = \gamma_2$ and $\gamma_2^\prime = \gamma$ so that the effective $R_{\text{eff}}$ is odd under R1 and R2. But the projection of the vector potential $A (A_1 | A_2)$ on the bonds under study in the polarisation $(a'b')$ implies $\gamma_1 = -\gamma_2$, $\gamma_1' = -\gamma_2'$, $\gamma_1'' = \gamma\cos(2\alpha)$ (\gamma microscopic constant, $\alpha$ angle in Fig. 1), i.e. an even component implying $R_{\text{eff}} = 0$. On the contrary, polarisation $(a'b')$ yields $\gamma_1 = -\gamma_2 = -\gamma_1' = -\gamma_2'$ so that $R_{\text{eff}} \neq 0$. This finding agrees nicely with experiment where on $T \to 0$ the intensities almost vanish for $(a'b')$ but grow for $(a'b')^\prime$. Additionally, $\gamma_1 = -\gamma_2$ and $\gamma_2 = -\gamma_2'$ imply odd parity under SD so that $R_{\text{eff}}$ is indeed $\Gamma_3$, not $\Gamma_4$. Calculating the next $\Gamma_3, S = 0$ bound state (less systematically) yields $45\text{cm}^{-1}$ in good agreement with the experimental $46\text{cm}^{-1}$ 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 interplan coupling $J_{1\perp}$ which is fully frustrated not changing the dimer spins [2]. We have employed a Dlog-Padé approximant for the $1/T$ expansion of the 2D $\chi_{\text{2D}}$ [10] complemented by the condition $\Delta = 2.98\text{meV}$. This ansatz works fine for $T > 35\text{K}$. The 3D $\chi_{\text{3D}}$ is computed from $\chi_{\text{2D}}$ on the mean-field level as $\chi_{\text{3D}}^{-1} = \chi_{\text{2D}}^{-1} + 4J_{1\perp}$. The inset in Fig. 4 shows that theory ($J_{1\perp} = 1.3(2)\text{meV}$) and experiment [1] agree without flaw above $40\text{K}$. Our value for $J_{1\perp}$ is significantly higher than the one in Ref. [2] 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 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 analyse the Shastry-Sutherland lattice as realized in SrCu$_2$(BO$_3$)$_2$. To our knowledge it is the first quantitative description of 2-particle bound states in 2D.

The symmetries of experimentally relevant states were determined. The reliability of the high order results allows to fix the experimental coupling constants very precisely ($J_1 = 6.16(10)\text{meV}$, $J_2/J_1 = 0.603(3)$, $J_3 = 1.3(2)\text{meV}$). Thereby, different experiments (ESR, FIR, INS, 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 $S = 1$ 3-triplet bound state is found at $x \approx 0.63$ indicating a transition to a triplet condensate probably related to the helical phase found previously [24,11]. We conjecture that this transition is first order occurring at lower $x$ than assumed so far. 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 phase beyond.

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