SrCu$_2$(BO$_3$)$_2$ – a Two Dimensional Spin Liquid

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We study an extended Shastry-Sutherland model for SrCu$_2$(BO$_3$)$_2$ and analyze the low lying parts of the energy spectrum by means of a perturbative unitary transformation based on flow equations. The derivation of the 1-magnon dispersion (elementary triplets) is discussed. Additionally, we give a quantitative description (symmetries and energies) of bound states made from two elementary triplets. Our high order results allow to fix the model parameters for SrCu$_2$(BO$_3$)$_2$ precisely: $J_1 = 6.16(10)$meV, $x := J_2/J_1 = 0.603(3)$, $J_3 = 1.3(2)$meV. To our knowledge this is the first quantitative treatment of bound states in a true 2d model.

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Presently low dimensional quantum antiferromagnets are investigated intensively both in experiment and theory. Systems that do not show a long ranged ordered ground state, so called spin liquids, are particularly interesting. Besides low spin and low coordination number spin liquids are more likely to form in strongly frustrated geometries. The recently synthesized antiferromagnetic material SrCu$_2$(BO$_3$)$_2$ [3,4] (cf. Fig. 1) is a nice two dimensional example since it is a realization of the Shastry-Sutherland model [3].

![Diagram](1) A part of the extended Shastry-Sutherland model. The circles are $S = 1/2$ entities. For $J_3 = 0$ this model reduces to the original Shastry-Sutherland model. The coupling $J_1$ is assumed to be antiferromagnetic. The starting point of our analysis is the limit of strong dimerization ($J_2, J_3 \rightarrow 0$).

In this model frustration plays an essential role. Each spin is coupled to pairs of spins on dimers. If these pairs form singlets the couplings between dimers is without effect and the singlet-on-dimers product state is always an eigen state and for certain parameters the ground state.

In this **diner phase** the system is gaped. The Hamiltonian reads

$$H = J_1 \sum_{<i,j>} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{<i,k>} \vec{S}_i \cdot \vec{S}_k + J_3 \sum_{<i,l>} \vec{S}_i \cdot \vec{S}_l,$$

where the bonds corresponding to interactions $J_1$, $J_2$ and $J_3$ are shown in Fig. 1. For larger ratios $x = J_2/J_1$ and $y = J_3/J_2$ the system transits into a Néel phase, competing with a helical phase for certain values of $y$. Finally there exists a ferromagnetic phase in the region of larger negative $x$ and $y$. A profound analysis of these scenarios can be found in Ref. [3]. Along the $x$-axis Koga and Kawakami [3] found an additional plaquette-singlet phase, which we see as being related to the helical phase.

In this article we start from the dimer phase where an elementary excitation is given by breaking up one singlet and substituting a triplet instead, which acquires a dispersion by hopping from dimer to dimer. For the next higher excitations we focus on bound states formed from pairs of the elementary triplets. We will perturbatively calculate the low lying excitations of the model about the limit of strong dimerization. The problem can be stated as

$$\frac{H}{J_1} = H_0 + xH_S, \quad \text{with } H_S = H_1 + \frac{y}{x} H_2 .$$

In the limit of isolated dimers ($x = 0$, with $y/x$ finite) $H$ is bounded from below and has an equidistant energy spectrum, since $H_0$ simply counts the number of excited dimers (up to a trivial constant). Furthermore $H_S$ can be decomposed

$$H_S = T_{-1} + T_0 + T_1 ,$$

where $T_i$ creates $i$ (destroys for $i < 0$) elementary triplets (energy quanta). These properties of $H_0$ and $H_S$ allow us to use the perturbative unitary transformation [6] based on flow equations [10]. This technique enables us to link smoothly and uniquely $H$ at $x \neq 0$ to an effective $H_{\text{eff}}$ conserving the number of triplets on dimers $[H_{\text{eff}}, H_0] = 0$. This permits a clear distinction between the ground state (without triplets), the 1-triplet sector, the 2-triplet sector etc.. Thus the effective Hamiltonian is block diagonal. The explicit form is given by

$$H_{\text{eff}} = H_0 + \sum_{k=1}^{\infty} x^k \sum_{|m|=k,M(m)=0} C(m)T(m) \cdot$$

$$\sum_{|m|=k,M(m)=0} C(m)T(m) .$$

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where $m$ is a vector of dimension $k$ of which the components are in $\{\pm 1, 0\}$; $M(m) = 0$ signifies that the sum of the components vanishes which reflects the conservation of the number of energy quanta. The operator product $T(m)$ is defined by $T(m) = T_{m_1}T_{m_2}\cdots T_{m_k}$. The coefficients $C(m)$ are generally valid fractions, which we computed up to order $k = 15$.

In terms of $H_{\text{eff}}$ it is easy to show that a hopping, displacing an elementary triplet by a finite distance, starts in sixth order in $x$, while in the 2-triplet sector correlated hopping starts in second order (see [1] and Refs. therein), explaining the rather flat 1-triplet dispersion in contrast to the much stronger pronounced 2-triplet dispersion as found by Kageyama et al. by INS measurements [12]. By means of degenerate perturbation theory Momoi and Totsuka [13] also derive an effective Hamiltonian for the Shastry-Sutherland model. Their third order result also shows the significance of correlated hopping in the multi-triplet dynamics of the model.

Let $|r\rangle = |r_1, r_2\rangle$ denote the state of the system with one triplet on the dimer at $r$ and singlets on all other sites. The amplitude $t^{o(r)}_{r'\rightarrow r}$ for a triplet-hopping from site $r$ to site $r'$ is then given by

$$t^{o(r)}_{r'\rightarrow r} = \langle r' | H_{\text{eff}} | r \rangle ,$$  

where the upper index $o(r) \in \{v, h\}$ allows to distinguish whether the hopping started on a vertically oriented ($v$) or a horizontally oriented dimer ($h$). Further we choose to split the hopping amplitudes into a net part $t_n$ and a deviation part $d_n$ ($s = r' - r$)

$$t^{o(r)}_{n} = \bar{t}_{n} + e^{iQr}d_{n} ,$$  

with $Q = (\pi, \pi)$.

Since $H_{\text{eff}}$ conserves the number of triplets one has

$$H_{\text{eff}} | r \rangle = \sum_{r'} t^{o(r)}_{r'\rightarrow r} | r + r' \rangle .$$

We introduce the Fourier transformed states

$$|\sigma, k\rangle = \frac{1}{\sqrt{N}} \sum_{r} | r \rangle e^{i(k+\sigma)Qr}$$

with the total number of dimers $N$, the new quantum number $\sigma \in \{0, 1\}$ reflecting the sub lattice structure and $k$ a vector in the magnetic Brillouin zone (MBZ). $H_{\text{eff}}$ acts as a $2 \times 2$ matrix on the states $|\sigma, k\rangle$ and $|1 - \sigma, k\rangle$. Its diagonalization yields

$$\omega_{1/2}(k) = \frac{a_{\sigma} + a_{1 - \sigma}}{2} = \frac{1}{2} \sqrt{(a_{0} - a_{1})^{2} + 4b^2} .$$

Here we have defined

$$a_{\sigma} = \left[ \bar{t}_{0} + 2 \sum_{r>0} \bar{t}_{r} \cos((k + \sigma Q)r) \right] \text{ and}$$

$$b = 2 \sum_{r>0} \bar{d}_{r} \cos(kr) ,$$  

(10)

with $r > 0$ if $r_1 > 0$, or $r_1 = 0$ but $r_2 > 0$. Thus the 1-triplet dispersion splits into two branches. We want to point out, however, that at $k = 0$ and on the borders of the MBZ the two branches fall onto each other leading to a 2-fold degenerate dispersion at these points. This can be derived by showing that the square root in Eq. (6) vanishes at these points [14]. An analogous degeneracy is noticed in the 2-triplet sector. In Ref. [11][14] we give a detailed analysis of the symmetries of the model (2d space group p4mm with underlying point group 4mm) and show that the degeneracies are due to glide line symmetries.

We calculated the amplitudes $\bar{t}_{r}$ and $\bar{d}_{r}$ (and therefore the dispersion) as exact polynomials in $J_{1}, x$ and $y$ up to and including 15th order. Expanding the square root in Eq. (6) about the limit of vanishing $x$ and $y$ produces terms $\propto x^\alpha y^\beta$ with $\alpha + \beta \geq 10$. Hence the energy splitting starts in 10th order only. It is negligible for all reasonable values of $x$ and $y$.

By substituting $y = 0$ in $\omega_{0}(k)$ we verify the decimal numbers Weihong et al. [15] obtained previously. For three different sets of $J_{1}, x$ and $y$ the 1-triplet dispersion is plotted in the inset of Fig. 2. ESR [16], FIR [17] and INS [12] data suggest a value of $\omega(0) = 2.98$ meV. At finite $k$ we have to rely on the INS measurement, which have rather large errors. We get a very good agreement. But it is not possible to fix the model parameters unambiguously from information in the 1-triplet sector alone. Thus we continue our analysis in the 2-triplet sector.

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 $W_{V_{k\pm d,r,d'}} (W_{V_{\pm d,r,d}})$ starting with one triplet on a horizontal (vertical) dimer and another at distance $d$. The action of $H_{\text{eff}}$ is to shift the triplets to $r$ and to $r + d'$. 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 $d, d' > 0$, because the exchange parity is fixed. The coefficients $W$ for $S \in \{0, 1, 2\}$ and the 1-triplet hopping yield the complete 2-particle dynamics. We compute $W$ up to $x^{12}$, the coefficients for the lowest-lying states even up to $x^{14}$.

Analogously to the 1-triplet problem we use the following basis for the 2-triplet states

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

(11)

where $k$ is the conserved total momentum in the MBZ applying due to the two sub lattices; $|r, r + d\rangle$ denotes
the state with triplets at \( \mathbf{r} \) and at \( \mathbf{r} + \mathbf{d} \) (\( \mathbf{d} > 0 \)). Since 1-triplet hopping is of higher order than the 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 leading 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 \( 8 \times 8 \) or \( 24 \times 24 \) matrix has to be analyzed. 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.

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 [18]. The dispersion of bound states starts only in \( x^3 \) (contrary to \( x^4 \) claimed in Ref. [19]). 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. 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.

**FIG. 2.** Energy of the lowest-lying \( S = 0 \) states. Curves refer to \( \mathbf{k} = 0 \) except the dashed-dotted one. The dotted curve displays the continuum at \( 2\Delta \). Inset: 1-triplet dispersion for various values of \((J_1/\text{meV}; x; y)\): (6.56;0.615;0) dashed-dotted line, (6.67;0.59;0.05) dashed line, (6.16;0.603;0) solid line.

The extrapolated energies are depicted in Figs. 3 (\( S = 0 \)) and 4 (\( S = 1 \)) as functions of \( x \). The double degeneracy for \( \mathbf{k} = (0, \pi) \) results from the same symmetry reasons as described for the 1-magnon dispersion. 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. [4]). Only above \( x \approx 0.55 \) the relation \( \omega(0) < \omega((0, \pi)) \) holds for \( S = 1 \).

**FIG. 3.** As in Fig. 2 for \( S = 1 \). Inset: Magnetic susceptibility. Theory (dashed) with directional rms average \( y = 2.13 \) [16], \( x, J_1 \) as in Fig. 2 experiment (solid) on powder [3].

For \( S = 0 \), the lowest mode vanishes at the same \( x \) as does the elementary triplet gap \( \Delta \). So no additional instability occurs for \( S = 0 \). This provides evidence against the competing singlet phase as presumed in Ref. [6]. 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 \( S = 0 \) (discussed in more detail in Ref. [14]). Assuming \( \Delta = 2.98 \text{meV} \) as above and \( \omega|_{S=1} = 4.7 \text{meV} \) from ESR [17], FIR [18] and INS [19] we can determine the model parameters for \( \text{SrCu}_2(\text{BO}_3)_2 \) precisely (\( y = 0 \)): \( x = 0.603(3) \) and \( J_1 = 6.16(10) \text{meV} \).

The inset in Fig. 2 shows that the 1-magnon dispersion agrees very nicely with experiment for these values. Further, the energy of the \( \Gamma_3 \) singlet matches the 30cm\(^{-1}\) peak in Raman scattering [14] perfectly. The \( \Gamma_4 \) singlet at 25cm\(^{-1}\) is forbidden by symmetry, since the Raman operator is \( \Gamma_3 \) at \( T = 0 \) [11]. Calculating the next \( \Gamma_3 \), \( S = 0 \) bound state (not shown) yields 45cm\(^{-1}\) in good agreement with the experimental 46cm\(^{-1}\) line, too.

We conclude that the 2D model (Fig. 4) explains the low-lying excitations of \( \text{SrCu}_2(\text{BO}_3)_2 \) perfectly. Thermodynamic quantities like the susceptibility \( \chi(T) \) require the inclusion of the interplane coupling \( J_\perp \) which is fully frustrated not changing the dimer spins [20]. The 3D \( \chi_{3D} \) is computed from \( \chi_{2D} \) on the mean-field level as
\( \chi_1 - 3_{3D} = \chi_1 - 3_{2D} + 4 J_{\perp} \) (inset in Fig. 3). Fits to the experiment [2] give \( J_{\perp} = 1.3(2) \text{meV} \) leading to a curve agreeing without flaw above 40K. Our value for \( J_{\perp} \) is significantly higher than the one in Ref. [20] due to different values of \( x \) and \( J_1 \).

Summarizing, we presented the first quantitative description of 2-particle bound states in 2D.

An unexpected instability for the \( S = 1/2 \)-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 [21,7]. We conjecture that this transition is first order occurring at lower \( x \) than assumed so far.

The symmetries of experimentally relevant states were determined. The reliability of the high order results allows to fix the experimental coupling constants very precisely (\( J_1 = 6.16(10) \text{meV}, J_2/J_1 = 0.603(3), J_{\perp} = 1.3(2) \text{meV} \)). Thereby, various experiments (ESR, FIR, INS, Raman, \( \chi(T) \)) are explained consistently.

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