Dynamic Structure Factor of the Two-Dimensional Shastry-Sutherland Model

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We calculate the 2-triplon contribution to the dynamic structure factor of the 2-dimensional Shastry-Sutherland model, realized in SrCu$_2$(BO$_3$)$_2$, by means of perturbative continuous unitary transformations. For realistic parameters we find flat bound 2-triplon bands. These bands show large weight in the structure factor depending strongly on momentum. So our findings permit a quantitative understanding of high precision inelastic neutron scattering experiments.

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Quantum antiferromagnets which do not have a long range ordered ground state, so-called spin liquids, continue to attract considerable interest. While there are many 1-dimensional examples there are only a few 2-dimensional systems. Of strong recent interest is SrCu$_2$(BO$_3$)$_2$ [1], a realization of the 2-dimensional spin 1/2 Shastry-Sutherland model [2]

$$H = J_1 \sum_{<i,j>} S_i S_j + J_2 \sum_{[i,j]} S_i S_j = J_1 \hat{U} + J_2 \hat{V},$$

(1)

where $J_1$ and $J_2$ are the intra- and inter-dimer couplings as in Fig. 1. Two spins coupled by $J_1$ are referred to as dimers. We focus on $J_1, J_2 > 0$.

The state $|0\rangle$ with singlets on all dimers is an exact eigen-state of $H$ for all values of $J_1$ and $J_2$ [2]. We found $|0\rangle$ to be the ground state (singlet-dimer phase) of $H$ for $x := J_2/J_1$ below $\approx 0.63$ [3] while other results indicate an instability at slightly higher values of $x$, for a review see Ref. [4].

In many papers it has been shown that the magnetic properties of SrCu$_2$(BO$_3$)$_2$ can be understood well by $H$ in (1) in the singlet-dimer phase [4]. Thus the bo rate constitutes a particularly transparent case of a 2-dimensional spin liquid. In view of the extensive spectroscopic data on this system quantitative theoretical results for spectral densities are highly desirable. But so far only the numerical exact diagonalization (ED) for systems of 20 or 24 spins was possible [4]. This approach is hampered by the finite size in two ways. First, the energies of the excited states display strong finite size effects since these states are spatially extended, in particular the bound states built from two elementary excitations. Second, the ED provides only isolated spikes instead of continuous distributions. In this article, we remedy these drawbacks by making use of recent conceptual progress in the method of perturbative continuous unitary transformations (CUTs) [5, 6]. We provide high order results for the dynamic structure factor which is measured by inelastic neutron scattering (INS).

We briefly review the derivation of perturbative effective operators (Hamiltonian and observable) in general and derive the appropriate INS observable for the Shastry-Sutherland model. The corresponding spectral density, i.e., the dynamic structure factor, is calculated via the $T = 0$ Green function. We focus on the 2-triplon part above the flat, featureless 1-triplon band [7–9].

The limit of isolated dimers, i.e., $x = 0$, serves as starting point of the perturbative analysis. The basic excitation is given by promoting one singlet to a triplet. The next higher excitation are two triplets and so on. Upon switching on the inter-dimer coupling ($x > 0$) the triplets acquire a dispersion and become dressed particles, which we call triplons [10].

Up to a constant, $\hat{U}$ in (1) counts the number of triplons and we define the particle-number operator $Q = \hat{U} + 3N/4$ ($N$: number of dimers). The perturbative part $\hat{V}$ in (1) decomposes into ladder operators $\hat{V} = T_{-1} + T_0 + T_1$, where the index $i$ denotes the number of triplons created (destroyed) by $T_i$.

The original Hamiltonian is mapped by a perturbative CUT [3, 9, 11] to an effective Hamiltonian (in units of $J_1$)

$$H_{\text{eff}}(x) = \hat{U} + \sum_{k=1}^{\infty} x^k \sum_{m} C(m)T(m)$$

(2)
where \( m = (m_1, m_2, \ldots, m_k), m_i \in \{0, \pm 1\} \). This effective Hamiltonian conserves the number of triplons: \[ H_{\text{eff}}, Q = 0 \] in each order \( k, H_{\text{eff}} \) is a sum of virtual processes \( T(m) = T_{m_1} \cdots T_{m_k} \) weighted by rational coefficients \( C \). The sum \( \sum_k \) is restricted by the triplon-conservation condition \( m_1 + \cdots + m_k = 0 \). The effective Hamiltonian can be decomposed into irreducible n-particle operators \( H_n \) of \( H_{\text{eff}} = H_0 + H_1 + H_2 + \ldots \).

The matrix elements of the irreducible \( H_n \) for the infinite system can be computed perturbatively on finite clusters due to the linked cluster theorem. For the Shastry-Sutherland model \( H_0 \) is conveniently set to zero; \( H_1 \) and \( H_2 \) were determined previously \([3, 9]\) to obtain the 1- and 2-triplon energies.

Applying the same transformation as for \( H \) other observables are also mapped onto their effective counterparts \([5]\)

\[
O_{\text{eff}}(x) = \sum_{k=0}^{\infty} x^k \sum_{i=1}^{k+1} \sum_{|m|=k} \tilde{C}(m;i)O(m;i) \\
O(m;i) := T_{m_1} \cdots T_{m_{i-1}} OT_{m_i} \cdots T_{m_k},
\]

where \( O \) is the initial observable. A useful decomposition of \( O_{\text{eff}} \) reads

\[
O_{\text{eff}} = \sum_{n=0}^{\infty} \sum_{d \geq -n} O_{d,n},
\]

where \( d \) indicates how many particles are created \((d \geq 0)\) or destroyed \((d < 0)\) by \( O_{d,n} \) whereas \( n \geq 0 \) is the minimum number of particles that must be present for \( O_{d,n} \) to have a non zero action. For \( T = 0 \) measurements only the \( O_{d,0} \) operators matter.

The energy and momentum resolved n-particle spectral density for the operator \( O \) is given by

\[
S_n^{(n)}(\omega, K) = -\pi^{-1} \text{Im} G^{(n)}(\omega, K),
\]

where \( G^{(n)} \) is the retarded n-particle Green function

\[
G^{(n)}(\omega, K) = \left\langle 0 \left| O_{n,0}^\dagger \frac{\omega - \sum_{i=1}^n H_{n,i} + i0^+}{\omega - \sum_{i=1}^n H_{n,i}} O_{n,0} \right| 0 \right\rangle.
\]

Since expectation values do not change under unitary transformations the Green function (6) is not altered if the the effective operators are substituted for the initial ones. Using the decompositions of \( H \) and of \( O \) as well as the conservation of triplons by \( H_{\text{eff}} \) the individual sectors of different triplon numbers can be analyzed separately.

We focus on the 2-triplon sector and introduce the 2-triplon momentum states \([3, 12]\)

\[
|\sigma, K, d\rangle^S = \frac{1}{\sqrt{N}} \sum_r \text{e}^{i(K+\sigma Q)(|r, r+d|/2)} |r, r + d\rangle^S ,
\]

where \( S \in \{0, 1, 2\} \) is the total spin of the two triplons and \(|r, r + d\rangle \) is the state of one triplon on the dimer at \( r \) and the other one on the dimer at \( r + d \). The primitive vectors \( a \) and \( b \) span the dimer-lattice \( \Gamma_{\text{eff}} \) (Fig. 1). The vector \( K + \sigma Q \) lies within the (first) Brillouin zone (BZ) of the dual lattice \( \Gamma_{\text{eff}} \) spanned by the vectors \( a^* \) and \( b^* \); as usual \( Q = (\pi, \pi) \) in units of the inverse dimer-lattice constant. The additional quantum number \( \sigma \in \{0, 1\} \) is chosen such that \( K \) lies within the magnetic Brillouin zone (MBZ) which is the (first) Brillouin zone of the dual lattice \( \Gamma_{\text{AB}} \). The exchange parity of the two triplons is fixed by \( [\sigma, K, d]^S = (-1)^{n} [\sigma, K, -d]^S \), hence we restrict to \( d = (d_1, d_2) > 0 \) or \( (d_1 = 0 \text{ and } d_2 > 0) \).

For fixed total momentum \( K, H_1 \) of order 15 is chosen such that \( K \) lies within the magnetic Brillouin zone (MBZ) which is the (first) Brillouin zone of the dual lattice \( \Gamma_{\text{AB}} \). The exchange parity of the two triplons is fixed by \( [\sigma, K, d]^S = (-1)^{n} [\sigma, K, -d]^S \), hence we restrict to \( d = (d_1, d_2) > 0 \) or \( (d_1 = 0 \text{ and } d_2 > 0) \).

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We turn to analyzing the appropriate observable for the INS experiment on the SrCu\(_2\)(BO\(_3\))\(_2\). It reads \( F(q) = \sum_r S^r(x) e^{iq \cdot x} \), where \( S^r(x) \) is the \( z \)-component of the spin at the position \( x \) (dots in Fig. 1). The \( x \) must not be confused with the vectors \( r \in \Gamma_{\text{eff}} \) which denote the positions of the dimer centers.

The momentum transfer \( q \) measured in experiment is any vector in the dual space whereas the excitations of the Shastry-Sutherland model are labeled best by the momenta \( K \in \text{MBZ} \). The usual backfolding implies \( K(q) = q \mod (\Gamma_{\text{AB}}^*) \) and \( K(q) + \sigma(q)Q = q \mod (\Gamma_{\text{eff}}^*) \) whence \( \sigma(q) = 0 \) for \( q \in \text{MBZ} \) \mod (\Gamma_{\text{eff}}^*) \) and \( \sigma(q) = 1 \) otherwise.

We construct an operator \( N \) defined for \( K \) and \( r \) such, that \( N(K; r) \) and \( F(q; x) \) have the same action on the ground state \( |0\rangle \). The operator \( N \) will then be used to obtain the effective operator. It is a crucial feature particular to the Shastry-Sutherland model that the triplon vacuum \( |0\rangle \) is not changed by the CUT since it is an exact eigen-state. With a suitable convention for the singlet orientation the action of \( F \) on \( |0\rangle \) is

\[
F(q)|0\rangle = i \sin(q \cdot \delta_r) \sum_{r'} e^{i q \cdot r'} |r'\rangle + i \sin(q \cdot \delta_h) \sum_{r'} e^{i q \cdot r'} |r'\rangle .
\]

The sums run over all vertical dimers \( r^v \) and horizontal dimers \( r^h \). A state \( |r\rangle \) is defined by one triplon with \( S^z = 0 \) on the dimer at \( r \) and singlets elsewhere. The vectors \( \delta_{r/v} \) are defined in Fig. 1.

The appropriate local operator \( N(r) \) using the distances \( r \in \Gamma_{\text{eff}} \) reads \( N(r) = S^v(r) - S^h(r) \), where the subscripts 0 and 1 distinguish the two spins on dimer \( r \) such that \( N(r)|0\rangle = |r\rangle \). The momentum space representation is given by \( \sigma = 1 - \sigma \)

\[
N(q) = a(q)N(\sigma, K) + b(q)N(\sigma, K)|_{\sigma(q), K(q)} \]

\[
N(\sigma, K) = \sum_r e^{i (K+\sigma Q) \cdot r} N(r)
\]

\[
a(q) = i [\sin(q \cdot \delta_v) + \sin(q \cdot \delta_h)] / 2
\]

\[
b(q) = i [\sin(q \cdot \delta_v) - \sin(q \cdot \delta_h)] / 2 .
\]
which ensures $\mathcal{N}(q)|0\rangle = \mathcal{F}(q)|0\rangle$ for all $q$.

Including the microscopic details for SrCu$_2$(BO$_3$)$_2$ (Fig. 1) and denoting $q$ by $q = \hbar a^+ + \hbar b^+$ fixes the scalar products to $q_0\delta_{hk} = 0.717(h-k)$ and $q_1\delta_{hk} = 0.717(h+k)$. This completes the derivation of the excitation operator $\mathcal{N}(q)$ for the INS with momentum transfer $q$.

The action of the effective local operator $\mathcal{N}_{\text{eff}}(r)$ from Eq. (3) on $|0\rangle$ is implemented on a computer. Although $\mathcal{N}(r)$ exclusively produces 1-triplon states when acting on $|0\rangle$, $\mathcal{N}_{\text{eff}}(r)$ leads to states containing an arbitrary number of triplons. Focusing here on the 2-triplon channel we have to deal with $\mathcal{N}_{2,0}$. The calculations of the amplitudes of $\mathcal{N}_{2,0}$ can be performed on finite clusters, see Refs. [6, 12].

By substituting $\mathcal{N}_{2,0}(q)$ for $\mathcal{N}(r)$ in Eq. (9b) we obtain $\mathcal{N}_{2,0}(\sigma, K(q), d)$ that are used for the effective Hamiltonian. The 2-triplon amplitudes $A_{\sigma, K, d} = \langle \sigma, K, d|\mathcal{N}_{2,0}(q)|0\rangle$ with $K \in$ MBZ define a vector in the quantum numbers $\sigma$ and $d$. Each component is calculated to $8^{th}$ order in $x$.

The 2-triplon energy and momentum resolved spectral density of $\mathcal{N}(\omega)$ is obtained by evaluating the 2-particle Green function (6) via tridiagonalization [13, 14]

$$
G^{(2)}(\omega, K, x) = \frac{\sum_{\sigma, d}|A_{\sigma, K, d}(x)|^2}{\omega - a_0 - \frac{b_1^2}{\omega - a_1 - \frac{b_2^2}{\omega - \ldots}}}.
$$

(10)

For fixed $K$, the continued fraction coefficients $a_i$ and $b_i$ are obtained by repeated application of $H_1 + H_2$ (matrix in $d$ and $\sigma$) on the initial 2-particle momentum state $|f_0\rangle = \mathcal{N}_{2,0}(q)|0\rangle$ (vector in $d$ and $\sigma$). Prior to the evaluation we extrapolate the matrix elements of $H_1$ and $H_2$ and the amplitudes of $|f_0\rangle$ by optimized perturbation theory (OPT) introduced in Ref. [6]. The values used for the OPT parameter $\alpha_{\text{OPT}}$ are $-0.20$ for the elements of $H_1, 0.80$ for the elements of $H_2$, and $-0.25$ for the amplitudes of $\mathcal{N}$ [12].

In Fig. 2, the spectral densities of $\mathcal{N}_{2,0}$ are plotted for the two momenta $K = (0, 0)$ and $(\pi, \pi)$, which translate to $q = (h, k)$ as indicated. Results are shown for two sets of parameters. The set $x = 0.635, J_1 = 7.33$ meV was proposed from the analysis of the magnetic susceptibility $\chi(T)$ [15]. We proposed the set $x = 0.603, J_1 = 6.16$ meV previously [3] based on the analysis of excitation energies at $T = 0$. Clearly, the differences between the two sets matter. Both the positions and the weights of the curves differ from one set to the other. Comparing to high resolution INS data [16] we come to the conclusion that the parameters $x = 0.603, J_1 = 6.16$ meV fit significantly better, both concerning the positions and the weights of the peaks. So we favor this set of parameters. It is objected that $\chi(T)$ is not well described [4]. But $\chi(T)$ is also strongly influenced by the presence of interlayer coupling [3, 15] which is not known. So a definite conclusion on the basis of $\chi(T)$ alone is very difficult. On the contrary, ED data for the specific heat [15] indicates that lower values of $x$ fit better to experiment than larger ones, cf. the data for 16 spins.

A possible weakness in our analysis are the necessary extrapolations. In Ref. [3], we did not use OPT but Dlog Padé approximants which allow for power-law singularities. This led in a very robust way to the instability at $x \approx 0.63$. OPT does not allow for power-law singularities. Hence it leads to a smoother dependence on $x$. No instability occurs below 0.7 so that the precise position of the instability is still an open issue. We emphasize that in spite of the smoother OPT extrapolation the parameters $x = 0.603, J_1 = 6.16$ meV still yield a better agreement with experiment than the parameters $x = 0.635, J_1 = 7.33$ meV.

To understand the 2-triplon states throughout the BZ, we calculate the spectral density for 150 different momenta. Color-coding the intensities leads to Fig. 3 where we follow the experimentally traced path in dual space. The black lines are the most relevant eigen-energies of $H_1 + H_2$ extracted from the 84x84 matrix representing the full matrix of $H_2$ plus a part of $H_1$ at fixed $q$ and $x$. Enlarging this matrix from 84x84 to 112x112 does not lead to visible changes. The dashed lines mark the lower and upper bound of the 2-particle continuum derived from the 1-triplon dispersion [9]. The energy range depicted is chosen according to a recent high resolution INS measurement of SrCu$_2$(BO$_3$)$_2$ [16]. The 1-triplon contribution would appear as a sharp, flat and highly intensive (red) band at about 3 meV from which no new insight is gained.

![FIG. 2. Two-triplon contribution to the spectral densities of the observable $\mathcal{N}(q)$ representing the INS on SrCu$_2$(BO$_3$)$_2$ for two sets of parameters. For clarity, a broadening of 0.02$J_1$ is used. The inaccuracy of the peak positions due to the extrapolation is about 2.5%.](image-url)
Our results compare excellently with the experimental data. The main conclusion is that the rather flat bands of the 2-triplon states can indeed be understood. Previously, experiment [17] and theory [3, 18, 19] found evidence for significant correlated hopping of two triplons. So it came as a surprise that high resolution INS showed very flat features only. Previous results were limited in resolution (in momentum and in energy [17]), analysed only two points of the BZ [3] or were restricted to low values of $x$ [18, 19]. Fig. 3 shows that there are many bound states distributed over a fairly large energy range of about 1.5 meV. This range corresponds to the previous expectation of enhanced correlated hopping. But the smoothly connected eigen-energies do not display a significant dependence on momentum. We interpret this finding as evidence for level repulsion. Due to the negligible 1-triplon kinetic energy there is a relatively large number of individual states involved. Their energetic repulsion renders each individual band very flat.

We also computed the energy and momentum integrated weights. At $x = 0.603$, we find that about 50%+25% of the full weight, known from a straightforward sum rule, is covered by the 1- and 2-triplon excitations, respectively. The remaining 25% must be attributed to higher triplon-excitations. This finding agrees nicely with experiment, see e.g. the constant momentum scan in Fig. 2(a) of Ref. [17].

In conclusion, we like to stress three main results. (i) The scenario of strongly dispersing modes in the 2-triplon sector cannot be held up. We showed that these modes are also rather flat and we argue that this stems from level repulsion. (ii) The quantitative agreement with the high resolution INS [16] is very good. Not only the overall shape of the structure factor but also prominent details like the intensive flat modes at about 5 meV and the modes just below the continuum at 5.75 meV are reproduced. This observation supports our choice for the parameters $x = 0.603, J_1 = 6.16$ meV. (iii) Finally, we have demonstrated that perturbative CUTs are capable and well-suited to quantitatively calculate complex quantities like spectral densities also for two-dimensional models.

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