Instability of isolated triplet excitations on the Shastry-Sutherland lattice (SSL)

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Configurations of singlets and triplets on the SSL have been proposed in the literature as variational ground states of the Shastry-Sutherland model at fixed magnetization $M$. We prove, that isolated triplet excitations on the SSL are unstable if the coupling $\alpha$ falls below a critical value $\alpha_c \approx 2.0$. The instability should be visible in the compound SrCu$_2$(BO$_3$)$_2$ where a coupling $\alpha^* = 1.48$ is realized.

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INTRODUCTION

The discovery of plateaus in the magnetization curve $M = M(B)$ of the compound SrCu$_2$(BO$_3$)$_2$ at rational values of the magnetization $M/M_S = 1/3, 1/4, 1/8$ ($M_S = 1/2$) has led to intensive investigations of the ground state properties of the Shastry-Sutherland model with Hamiltonian

$$H = \sum_{(x,y)} S(x)S(y) + \alpha \sum_{(x,y)} S(x)S(y).$$  \hspace{1cm} (1)

The spin-1/2 couplings extend over nearest-neighbor (\langle x, y \rangle) and specific next-nearest-neighbor (\langle \langle x, y \rangle \rangle) couplings, which define the SSL, shown in Fig. 1.

![Diagram showing the couplings in the Shastry-Sutherland model. Nearest and next-nearest neighbor bonds are represented by dotted and solid lines, respectively. The diagonal bonds define the Shastry-Sutherland lattice (SSL).](image)

In the absence of an external field ($M=0$) the ground state can be represented by a product of singlet states on the SSL if $\alpha$ exceeds a critical value $\alpha_c$, which has been found to be $\alpha_c = 1.43$ in Ref. [5]. This value is very close to the coupling $\alpha^* = 1.48$, which seems to be realized in the compound SrCu$_2$(BO$_3$)$_2$. In order to describe the magnetic properties of the compound – in particular for fixed magnetizations $M = 1/4, 1/6, 1/8, 1/12, 1/16$ where plateaus are expected or found – several ordering patterns have been proposed and discussed in the literature. \hspace{1cm} \[ \hspace{1cm} \]

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They all have in common that in the sector with total spin $S$, $N_T = S$ triplets and $N_S = N/2 - N_T$ singlets are distributed over the SSL with $N$ sites. Typical examples of these singlet-triplet configurations are shown in Ref. [9]. In contrast to the pure singlet configuration (Fig. 1) the singlet-triplet configurations are not eigenstates of the Hamiltonian $H$. Moreover, it has been shown in our previous paper [10] that the formation of triplets on the SSL is disadvantageous if the coupling $\alpha$ falls below a certain critical value $\alpha_c(M)$, depending on $M$. In particular, the singlet-triplet configurations proposed in Fig. 8 of Ref. [10] cannot be considered as adequate variational states for $M = 1/8$, since $\alpha_c(M = 1/8) = 2.3 > \alpha^*$. As an alternative, we proposed in Ref. [10] (Fig. 4) a “monomer-dimer configuration” built up from monomers (i.e. isolated spin-up states) and dimers (i.e. singlets) on nearest and next-nearest neighbor bonds.

It is the purpose of this Letter, to prove that the instability of isolated triplet excitations on the SSL is a quite general feature for $\alpha < 2$ and low magnetizations $M \leq 1/8$.

SINGLET-TRIPLET VERSUS MONOMER-DIMER CONFIGURATIONS

The variational states $|K, \nu\rangle$ which are constructed from the monomer-dimer configurations generalize the familiar singlet-triplet configurations on the SSL in the following sense: In the sector with total spin $S$ we have $\nu = 2S$ monomers – i.e. isolated spin-up states $|x_j\rangle$ at sites $x_1, \ldots, x_\nu$. They form the ferromagnetic cluster ($\nu$). All remaining sites $x, y, \ldots$ are occupied with $(N - \nu)/2$ singlets (dimers) $|x, y\rangle$. They form the antiferromagnetic cluster ($K$). Each monomer and dimer configuration defines a variational state:

$$|K, \nu\rangle = \prod_{j=1}^{\nu} |x_j\rangle + \prod_{(x,y) \in K} [x, y].$$  \hspace{1cm} (2)
The expectation value of Hamiltonian (1) between these states turns out to be [10]

$$\langle K, \nu | H | K, \nu \rangle = -\frac{3}{4} \left( N_1^{(0)}(K) + \alpha N_2^{(0)}(K) \right)$$

$$+ \frac{1}{4} \left( N_1^{(1)}(\nu) + \alpha N_2^{(1)}(\nu) \right)$$

(3)

where $N_1^{(0)}(K)$ and $N_1^{(1)}(\nu)$ are the numbers of nearest neighbor singlets on $(K)$ and monomer pairs on $(\nu)$, respectively. $N_2^{(0)}(K)$ and $N_2^{(1)}(\nu)$ denote the corresponding numbers on the next-nearest neighbors according to Fig. 3. The singlet-triplet configurations on the SSL are special monomer-dimer configurations with:

$$N_1^{(0)} = 0, \quad N_2^{(0)} = \frac{N - \nu}{2}, \quad N_1^{(1)} = 0, \quad N_2^{(1)} = \frac{\nu}{2}.$$  

(4)

i.e. we have a maximum number of dimers (singlets) and monomer pairs (triplets) on the SSL. Due to the geometry of the SSL it is not possible to maximize the number of singlets $N_2^{(0)}$ and to minimize the number of triplets $N_2^{(1)}$ at the same time. Here begins the problem with triplet excitations on the SSL: For smaller values of $\alpha$ [$(\alpha < \alpha_c(M)]$, the break up of the triplets into well separated monomer pairs is favored energetically. For $M \leq 1/8$ – where the triplets on the SSL are well isolated (cf. Figs. 8,9,10 in Ref. [3]) – we can study the break up of each triplet and the change of the local environment in the monomer-dimer configuration directly, as is shown in Fig. 2(a),(b). The configuration $(K_a, \nu_a)$ in Fig. 2(a) shows a triplet at sites $x$ and $y$ embedded in a sea of singlets on the SSL. They are represented by the solid lines in Fig. 2(a). The triplet is broken up in a pair of monomers at sites $x$ and $y$ separated by a “knight’s move” [cf. Fig. 2(b)]. Comparing the energies of the configurations $(K_a, \nu_a)$ and $(K_b, \nu_b)$ we have to decrease $N_2^{(0)}$ and $N_2^{(1)}$ and to increase $N_1^{(0)}$ by one unit:

$$\Delta E = \langle K_b, \nu_b | H | K_b, \nu_b \rangle - \langle K_a, \nu_a | H | K_a, \nu_a \rangle$$

$$= \frac{1}{2} \left( \alpha - \frac{3}{2} \right).$$

(5)

The energy difference (5) is negative for $\alpha < 3/2$. Therefore, the break up of the triplet is favored in this regime. We will see in the next section, that this phenomenon occurs already at larger values of $\alpha$, if we improve our variational ansatz.

**MAGNETIC ORDER AND THE FROZEN MONOMER APPROXIMATION**

The monomer-dimer configurations are not eigenstates of the Hamiltonian (1). However, it might happen that the ground state is governed by a specific distribution $(\nu)$ of “frozen” monomers. They define the magnetic ordering of the ground state. The variational ansatz (2) will be improved if we substitute the product of dimers

$$\prod_{(x,y) \in K} \langle x, y \rangle \rightarrow \Psi(K)$$

(6)

by the ground state $\Psi(K)$ of the antiferromagnetic cluster Hamiltonian $H(K)$:

$$H(K) \Psi(K) = E(K) \Psi(K).$$

(7)

$H(K)$ contains all nearest and next-nearest neighbor couplings on the antiferromagnetic cluster $K$. In Fig. 2(a),(b) they are represented by the solid singlet lines and the dotted lines connecting them.

The variational ansatz (2) yields an upper bound on the ground state energy $E_0(M = \nu/2N) \leq E(K, \nu)$ in the sector with magnetization $M$, where

$$E(K, \nu) = \frac{1}{4} \left( N_1^{(1)}(\nu) + \alpha N_2^{(1)}(\nu) \right) + E(K).$$

(8)

Concerning the triplet-singlet configuration in Fig. 2(a), the ground state $\Psi(K_a)$ of the antiferromagnetic cluster
Hamiltonian $H(K_a)$ is again given by the product of singlets on the SSL. Note that each singlet (e.g. $[1, 2]$) is accompanied by two nearest neighbor couplings $S(1)S(2), S(2)S(3)$ such that the total spin operator $S(1) + S(2)$ acts on the singlet $[1, 2]$. For this reason the ground state energy cannot be lowered in the case $(K_a, \nu_a)$.

The situation is different in configurations with isolated monomers. The ground state $\Psi(K_b)$ of the antiferromagnetic cluster is not identical with the dimer product shown in Fig. 2(b). If we apply here the couplings $S(2)S(3), S(4)S(5)$ and $S(4)S(7)$ onto the nearest neighbor singlet $[3, 4]$ a new state is created. Therefore, the ground state energy $E(K_b)$ of the antiferromagnetic cluster $K_b$ will be lower than the expectation value of the corresponding monomer-dimer configuration in Fig. 2(b).

This will shift the critical value $\alpha_c$ where the triplet excitation becomes unstable to larger values $\alpha_c > 3/2$. Indeed, we can derive an improved lower bound for $\alpha_c$, if we substitute the dimers $[1, 2][3, 4][5, 6][7, 8]$ by an antiferromagnetic cluster with 8 sites, as shown in Fig. 2(c). The ground state energy $E_8(\alpha)$ of the 8-site cluster is lower than the energy of the corresponding dimer configuration in Fig. 2(b) by an amount

$$\Delta_8(\alpha) = -\frac{3}{4}(3\alpha + 1) - E_8(\alpha); \quad (9)$$

$\Delta_8(\alpha)$ is shown in Fig. 2a. The straight line $\Delta E = (\alpha - 3/2)/2$ represents the energy difference $\Delta_8$. Therefore, the intersection point

$$\alpha_c = 1.95 \quad (10)$$

yields the critical value, where the instability of the triplet sets in. Enlarging the antiferromagnetic surrounding of the frozen monomer pair in Fig. 2(c) leads to an increase of the critical value $\alpha_c$. However, the effect is small: a calculation with a 16-site cluster containing the 8-site cluster yields $\alpha_c = 1.97$.

The changes in the magnetic order from an isolated triplet on the SSL to a pair of monomers separated by a “knight’s move” can be tested by measuring the spin-spin structure factors, which are obtained from spin-spin correlators $S_3(x)S_3(y)$.

The expectation values of the spin-spin correlators can be read off from the monomer-dimer configurations:

$$\langle K\nu|S_3(x)S_3(y)|K\nu\rangle = \frac{1}{4}\{\delta^{(1)}(x,y) - \delta^{(0)}(x,y)\} \quad (11)$$

where $\delta^{(1)}(x,y) = 1$ and $\delta^{(0)}(x,y) = 1$ only if $(x,y)$ coincides with nearest or next-nearest neighbor bonds on $(K, \nu)$ occupied with a monomer pair or a dimer, respectively. In all other cases $\delta^{(j)}(x,y) = 0$. Note in particular, that all correlators with one site $x$ on the ferromagnetic cluster $\nu$ and the other site $y$ on the antiferromagnetic cluster $K$ vanish.

In the frozen monomer approximation with an antiferromagnetic cluster wavefunction $\Psi(K)$, (11) can be extended to

$$\langle \Psi(K), \nu|S_3(x)S_3(y)|\Psi(K), \nu\rangle = \frac{1}{4}\{\delta^{(1)}(x,y) + \langle \Psi(K)|S_3(x)S_3(y)|\Psi(K)\rangle \delta(x,y)\} \quad (12)$$

Note that (11) and (12) differ in the correlations $S_3(x)S_3(y), x, y \in K$ on the antiferromagnetic cluster $K$.

![Diagram](image)

**FIG. 3:** The energy differences $\Delta_8, \Delta_{16}$ and $\Delta_{24}$. The intersection points define the critical coupling $\alpha_c$ where the break up of the triplet sets in.

**COEXISTENCE OF STABLE AND UNSTABLE TRIPLET EXCITATIONS ON THE SSL FOR $M = 1/6$**

The instability discussed so far only concerns “isolated” triplets on the SSL. The density of triplets $N_T/N = M$ is controlled by the magnetization $M$ and raises the question, what happens in the cases $M = 1/4$ and $M = 1/6$. A typical singlet-triplet configuration for $M = 1/4$ on the SSL is given in Fig. 6(a) of Ref. 1. Breaking up a triplet in these configurations produces additional monomer pairs, which increases the energy, i.e. a high density of triplets prevents their instability. In the case $M = 1/6$, the triplets on the SSL form stripes as is shown in Fig. 6(b). Interestingly enough, each second stripe of the triplets can break up as is demonstrated in Fig. 6(b). Comparing the energies of the configurations $(K_a, \nu_a)$ and $(K_b, \nu_b)$, we find again, that the energy difference $\Delta_8$ per triplet changes sign for $\alpha_c(M = 1/6) = 3/2$.

Note, that the isolated monomers along one stripe in Fig. 6(b) are accompanied with a quasi-one-dimensional antiferromagnetic cluster $K$. We have computed the ground state energy of the 24 site clusters $E_{24}(\alpha, M =
1/4), which surrounds the 4 pairs of monomers arising from the break up of 4 triplets on the SSL, shown in Fig. 4(b). The energy difference (per triplet)

$$\Delta_{24}(\alpha, 1/6) = \frac{1}{4} \left\{ -\frac{3}{4} (8\alpha + 4) - E_{24}(\alpha, 1/6) \right\} \quad (13)$$

is plotted in Fig. 3. It meets the straight line $(\alpha - 3/2)/2$ at

$$\alpha_c(M = 1/6) = 1.985 \quad (14)$$

This improved lower bound for $\alpha_c(M = 1/6)$ differs from the lower bound we found in Ref. [10]. There we did not realize, that a mixture of triplets on the SSL and isolated monomers – as shown in Fig. 4(b) – lowers the energy more efficiently.

![FIG. 4: (a) Singlet-triplet configuration ($K_a, \nu_a$) on the SSL for $M = 1/6$. (b) Break up of triplets along a stripe with an antiferromagnetic 24-site cluster ($K_b, \nu_b$).](image)

**DISCUSSION AND CONCLUSIONS**

In this Letter, we have investigated the stability of triplet excitations on the Shastry-Sutherland lattice. Our results can be summarized as follows:

(i) Isolated triplets turned out to be unstable if the ratio of next-nearest to nearest neighbor couplings is smaller than $\alpha < \alpha_c \simeq 2.0$. This implies that the magnetic properties of SrCu$_2$(BO$_3$)$_2$ – where the coupling ratio is $\alpha^* \simeq 1.48$ – cannot be understood in terms of isolated triplet excitations on the SSL, if the magnetization $M$ is small enough ($M \leq 1/8$). This statement holds in particular for the triplet ordering patterns for $M = 1/8, 1/12, 1/16$ in Figs. 8, 9, 10 of Ref. [9].

(ii) For $M = 1/6$ [Fig. 7(a) in Ref. [9]] the triplets form stripes on the SSL. It turns out that the triplets on each second stripe break up into isolated monomers if $\alpha \leq \alpha_c(M = 1/6) = 1.985$.

(iii) The triplets on the SSL in the configuration for $M = 1/4$ [Fig. 6(a) in Ref. [9]] appears to be stable for $\alpha = \alpha^*$. This demonstrates, that triplet excitations on the SSL are stabilized if the triplet density is high enough.

Breaking up the isolated triplet into two monomers (spin-up states) at sites $x$ and $y$ separated by a “knight’s move” [Fig. 2(c)] lowers the energy of the new configuration in two ways:

1. There is a “classical” effect, which results from the rearrangement of singlets and triplets on ($K_a, \nu_a$) [in Fig. 3(a)] into the monomer-dimer configuration ($K_b, \nu_b$) [in Fig. 3(b)].

2. There is a “quantum” effect produced by the antiferromagnetic cluster, which surrounds the monomers at sites $x$ and $y$ [Fig. 2(c)].

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