One-magnon states and electron spin resonance in spin-ladders with singlet-rung ground state in a staggered magnetic field

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Abstract

One-magnon problem for spin-ladders with exact singlet-rung ground state in arbitrary oriented constant and staggered magnetic fields is studied. For two special configurations when the both fields are parallel or perpendicular to each other the exact formulas for eigenstates and eigenvectors are presented. In the parallel case the one-magnon contribution to the ESR absorption line shape is obtained. The latter is non-Lorentzian, has finite range and a gap around resonance. For low fields both the range of the spectrum and the width of the gap are controlled by the width of the one-magnon zone in zero magnetic field. Some other interactions usually contributing to the ESR line shape are also briefly discussed.

1 Introduction

Effects of staggered magnetic field (SMF) in 1-D and 2-D spin systems attract now a considerable interest [1]. A violation of the rotational symmetry induced by SMF mixes the states with different polarizations while a violation of the translation symmetry mixes the states with wave numbers connected by the relation $k_2 = k_1 + \pi$. 
In the present paper we study these effects basing on the special spin-ladder model with exact singlet-rung ground state first suggested in [2]. An addition of rather small constant and staggered magnetic fields do not destroy the vacuum however the one-magnon problem transforms into the spectral problem for a special $6 \times 6$ matrix. When the both fields are parallel the remaining rotational invariance reduces the problem to three spectral problems for $2 \times 2$ matrices. As it will be shown in the Sect. 3 for the perpendicular configuration the problem also simplifies and reduces to two $3 \times 3$ problems.

Violation of translation symmetry in spin-ladders was discussed in [3] in connection with anomaly in Raman scattering in the ladder/chain system Sr$_{14}$Cu$_{24}$O$_{41}$. It was suggested that interaction constants of spin-ladder Hamiltonian are modulated by the ladder-chain interaction. Staggered magnetic field may be considered as a simple example of such modulation.

Electron spin resonance (ESR) is a traditional experimental test for small symmetry violations in spin systems [4], [5]. In the present model the ESR transitions do not change a number of magnons inducing only a change of polarization and a shift on $\pi$ of a wave number.

As it will be shown in Sect. 3 in the presence of SMF magnons with different polarizations have different dispersions. Owing to this fact the ESR line shape broadens. For parallel constant and staggered fields the exact formula for the one-magnon contribution to line shape is calculated in Sect. 4. The obtained line shape has a non-Lorentzian form, finite range and a gap around the resonance. In Sect. 5 we discuss briefly effects of some other symmetry violating terms such as dipole-dipole and Dzyaloshinskii-Morya interactions.

Spin gap systems such as spin-ladders and Haldane spin-chains have a significant feature. Their low-temperature properties with good precision may be obtained from analysis of a few number of low energy excitations [6], [7], [8]. Since the corresponding theoretical predictions may be tested by experiment, the spin-gap systems probably give an ideal possibility for theorists and experimenters to work in close connection. However in contrast to Haldane chains the ESR in spin-ladders was not studied so intensively. (See a very brief report in [9] devoted to ESR in CaV$_2$O$_5$). We believe that the present paper will stimulate an interest to ESR in spin-ladder materials and partially meet a lack in theoretical study of this subject mentioned in [4].
2 Spin Hamiltonian and magnon number operator

General spin-ladder Hamiltonian has the following form:

\[ \mathcal{H} = \sum_{n=-\infty}^{\infty} H_{n,n+1}^{(2)} + H_{n}^{(1)}, \]  

where

\[ H_{n,n+1}^{(2)} = H_{n,n+1}^{\text{leg}} + H_{n,n+1}^{\text{frust}} + H_{n,n+1}^{\text{cyc}}, \quad H_{n}^{(1)} = H_{n}^{\text{rung}} + H_{n}^{Z}, \]

and

\[ H_{n,n+1}^{\text{leg}} = J_{\parallel}(S_{1,n} \cdot S_{1,n+1} + S_{2,n} \cdot S_{2,n+1}), \]
\[ H_{n,n+1}^{\text{frust}} = J_{\text{frust}}(S_{1,n} \cdot S_{2,n+1} + S_{2,n} \cdot S_{1,n+1}), \]
\[ H_{n,n+1}^{\text{cyc}} = J_{c}(S_{1,n} \cdot S_{1,n+1})(S_{2,n} \cdot S_{2,n+1}) + (S_{1,n} \cdot S_{2,n})(S_{1,n+1} \cdot S_{2,n+1}) \]
\[ - (S_{1,n} \cdot S_{2,n})(S_{2,n} \cdot S_{1,n+1}), \]
\[ H_{n}^{\text{rung}} = J_{\perp} S_{1,n} \cdot S_{2,n}, \]
\[ H_{n}^{Z} = -g\mu_{B}(h + (-1)^{n}h_{st}) \cdot (S_{1,n}^{z} + S_{2,n}^{z}). \]

Here \( S_{i,n} \) \((i = 1, 2; n = -\infty...\infty)\) are spin-1\(^{2}\) operators associated with cites of the ladder, \( h \) and \( h_{st} \) are the constant and staggered magnetic fields. Except the Sect. 5 we shall suppose that \( h \) is oriented along the z-axis.

In this paper we shall restrict the Hamiltonian (1)-(3) by two additional conditions suggested in [2]. The first one is the following:

\[ [\mathcal{H}, \mathcal{Q}] = 0, \]

where \( \mathcal{Q} = \sum_{n} \frac{3}{4} I + S_{1,n} \cdot S_{2,n} \) is the magnon number operator or the sum of triplet-rung projectors.

Let us denote by \( |0\rangle_{n} \) and \( |1\rangle_{n}^{j} \) \((j = -1, 0, 1)\) the singlet and triplet states associated with \( n \)-th rung. Then the vector

\[ |0\rangle = \prod_{n} |0\rangle_{n}, \]

is the non degenerate zero-eigenstate of \( \mathcal{Q} \). According to (4) it is also an eigenstate of \( \mathcal{H} \). Our second condition demands the state (5) to be the ground state of \( \mathcal{H} \).

As it was first shown in [2] the condition (4) will be satisfied if \( J_{\text{frust}} = J_{\parallel} - \frac{1}{2} J_{c} \). The following inequalities \( J_{\perp} > 2J_{\parallel}, J_{\perp} > \frac{5}{2} J_{c}, J_{\perp} + J_{\parallel} > \frac{3}{4} J_{c} \) also guarantee that for rather small \( h \) and \( h_{st} \) the ground state has the form (5).
3 One-magnon states in staggered magnetic field

In the presence of SMF we have to suggest the following general form of a one-magnon state:

\[ |1, k; \nu\rangle_{\text{magn}} = \sum_{n=-\infty}^{\infty} \sum_{j=-1, 0, 1} \xi_j(k, \nu) e^{2i\kappa n} |1, 2n\rangle^j + \eta_j(k, \nu) e^{ik(2n+1)} |1, 2n + 1\rangle^j, \]

(6)

where \(-\frac{\pi}{2} \leq k \leq \frac{\pi}{2}\) and \(|1, n\rangle^j = \ldots |0\rangle_{n-1} |1\rangle_n |0\rangle_{n+1} \ldots\). The parameter \(\nu = -1, 0, 1\) enumerates magnon polarizations.

The Shrödinger equation for the amplitudes (6) is equivalent to the following spectral problem:

\[ A(k) \begin{pmatrix} \xi \\ \eta \end{pmatrix} = E(k, \nu) \begin{pmatrix} \xi \\ \eta \end{pmatrix}, \]

(7)

for the \(6 \times 6\) matrix

\[ A(k) = \begin{pmatrix} J_\perp - \frac{3}{2} J_c - g\mu_B (h \tilde{S}_z + h_{st} \cdot \tilde{S}) & J_c \cos k \\ J_c \cos k & J_\perp - \frac{3}{2} J_c - g\mu_B (h \tilde{S}_z - h_{st} \cdot \tilde{S}) \end{pmatrix}. \]

(8)

Here the \(3 \times 3\) matrices \(\tilde{S}\) represent the standard triple of \(S = 1\) spin operators.

For \(\mathbf{h}_{st} \parallel \mathbf{h}\) the problem (7) has the following system of solutions:

\[ \begin{pmatrix} \xi_+(k, \nu) \\ \eta_+(k, \nu) \\ \xi_-(k, \nu) \\ \eta_-(k, \nu) \end{pmatrix} = \begin{pmatrix} J_c \cos k \\ \nu g\mu_B h_{st} + \sqrt{J_c^2 \cos^2 k + \nu^2 g^2 \mu_B^2 h_{st}^2} \\ \nu g\mu_B h_{st} + \sqrt{J_c^2 \cos^2 k + \nu^2 g^2 \mu_B^2 h_{st}^2} \\ -J_c \cos k \end{pmatrix} \otimes e^\nu, \]

(9)

with

\[ E_\parallel(k, \nu) = J_\perp - \frac{3}{2} J_c - \nu g\mu_B h \pm \sqrt{J_c^2 \cos^2 k + \nu^2 g^2 \mu_B^2 h_{st}^2}, \]

(10)

where \(e^\nu \in \mathbb{C}^3\) and \(\tilde{S}^3 e^\nu = \nu e^\nu\) for \(\nu = -1, 0, 1\).

For \(J_c > 0\) in the limit \(h_{st} \to 0\) the solution \(\xi_+(k, \nu), \eta_+(k, \nu)\) turns into magnon with wave number \(k\) \([2]\) while the solution \(\xi_-(k, \nu), \eta_-(k, \nu)\) turns into magnon with wave number \(k + \pi\). For \(J_c < 0\) these limits change.

For \(\mathbf{h}_{st} \perp \mathbf{h}\) the action of the matrix \(A(k)\) decomposes the space \(\mathbb{C}^6\) on two invariant subspaces \(V_+^\perp\) and \(V_-^\perp\) generated by

\[ v_{\nu}^\perp(\pm) = \begin{pmatrix} e^\nu \\ \mp(-1)^\nu e^\nu \end{pmatrix}, \quad \nu = -1, 0, 1, \]

(11)
and the problem (7) splits on two problems for the matrices

\[ A_\pm^x(k) = (J_\perp - \frac{3}{2} J_c \pm \frac{1}{3} J_c \cos k) I - \mu_B h \hat{S}_z \pm 2 J_c \cos k (\hat{S}_z^2 - \frac{2}{3} I) - \mu_B h_{st} \hat{S}_x, \]  

(12)

where \( I \) is a unit matrix. Since \( A^x_\pm(k) = A^x_\pm(k + \pi) \) we may study only the ”+” problem in the interval \(-\pi < k \leq \pi\). After the following substitution

\[ E_\pm^x(k, \nu) = J_\perp - \frac{3}{2} J_c + \frac{1}{3} J_c \cos k + \frac{2}{3} J_c \varepsilon(k, \nu), \]  

(13)

the characteristic equation for the matrix \( A^x_+(k) \) transforms to the form

\[ \varepsilon^3(k, \nu) - 3(\cos^2 k + \frac{1}{3} \gamma^2 \gamma^2 (h^2 + h^2_{st})) \varepsilon(k, \nu) + 2 \cos k (\cos^2 k + \frac{1}{2} \gamma^2 (h^2_{st} - 2h^2)) = 0, \]  

(14)

where \( \gamma = \frac{3 \mu_B}{2 J_c} \). Now the standard substitution: \( \varepsilon(k, \nu) = \sqrt{\cos^2 k + \frac{1}{3} \gamma (h^2 + h^2_{st}) \lambda(k, \nu)} \), reduces this equation to the form \( \lambda^3(k, \nu) - 3 \lambda(k, \nu) + 2a(k) = 0 \) where

\[ a(k) = \frac{(\cos^2 k + \frac{1}{3} \gamma^2 (h^2_{st} - 2h^2)) \cos k}{\sqrt{(\cos^2 k + \frac{1}{3} \gamma^2 (h^2 + h^2_{st}))^3}}. \]  

(15)

It may be easily proved that \( |a(k)| \leq 1 \) and so putting \( a(k) = \sin \alpha(k) \), where \(-\frac{\pi}{6} \leq \alpha(k) \leq \frac{\pi}{6} \) we obtain the following representation for solutions of the Eq. (14)

\[ \varepsilon(k, \nu) = 2 \sqrt{\cos^2 k + \frac{1}{3} \gamma (h^2 + h^2_{st})} \sin (\frac{1}{3} \arcsin a(k) + \frac{2\pi}{3} \nu), \quad \nu = -1, 0, 1. \]  

(16)

The corresponding eigenvectors of the matrix \( A^x_+(k) \) may be represented as follows:

\[ v^\perp(\varepsilon(k, \nu)) = \begin{pmatrix} \frac{\sqrt{3}}{2} \gamma h_{st} (\varepsilon(k, \nu) - \cos k - \gamma h) \\ -\sqrt{3} \gamma h_{st} (\varepsilon(k, \nu) - \cos k - \gamma h) \\ \frac{\sqrt{3}}{2} \gamma h_{st} (\varepsilon(k, \nu) - \cos k + \gamma h) \end{pmatrix}. \]  

(17)

### 4 One-magnon ESR line shape for \( h_{st} \parallel h \)

According to [10] the ESR line shape depends on the imaginary part of magnetic susceptibility:

\[ \chi''(\omega, T) \propto \sum_{E_a > E_b} e^{-\frac{E_a + E_b}{2kT}} \sinh \frac{\omega}{2kT} |\langle a|S_x^{tot}|b\rangle|^2 \delta (E_a - E_b - \omega), \]  

(18)

where \( S_x^{tot} = \sum_n S_{1,n} + S_{2,n} \) is the total spin of the ladder. In this section we shall obtain \( \chi''(\omega) \) the one-magnon contribution to the formula (18). Calculation of \( \chi''(\omega) \) for generally
oriented $\mathbf{h}$ and $\mathbf{h}_{st}$ is very difficult even in the case $\mathbf{h} \perp \mathbf{h}_{st}$. However for $\mathbf{h}_{st} \parallel \mathbf{h}$ it may be easily developed by straightforward substitution of the formulas (9) and (10) into (18) (Of course with correct normalization account of the vectors (9)). The delta function removes summation in (18) and the main difficulty is to express correctly $\cos k$ from $\omega$.

The calculations show that all transitions are grouped in pairs. Contribution of each pair lies in the corresponding frequency interval and have one of the two possible forms,

$$\chi''_\pm(\omega, T) = \frac{\Gamma^2 \cosh \frac{1}{2kT}(\frac{\Gamma^2}{4(\omega + \omega_{res})} \pm \omega_{res})}{\Gamma^2 + (\omega \pm \omega_{res})^2} \sinh \frac{\omega}{2kT}, \tag{19}$$

where $\omega_{res} = g\mu_B h$ and $\frac{\Gamma}{2} = g\mu_B h_{st}$.

The two pairs $E^\parallel_+(k, 1) \rightarrow E^\parallel_+(k, 0)$, $E^\parallel_-^+(k, 0) \rightarrow E^\parallel_-^+(k, -1)$ and $E^\parallel_+(k, 0) \rightarrow E^\parallel_+(k, -1)$ contribute $\chi_-(\omega)$ in the frequency interval $\omega \leq \omega \leq \omega_{res} - \delta \omega$. The two pairs $E^\parallel_+(k, 1) \rightarrow E^\parallel_+(k, 0)$, $E^\parallel_-^+(k, 0) \rightarrow E^\parallel_-^+(k, -1)$ and $E^\parallel_+(k, 1) \rightarrow E^\parallel_+(k, 0)$, $E^\parallel_-(k, 0) \rightarrow E^\parallel_-(k, -1)$ also contribute $\chi_-(\omega)$ but in the frequency interval $\omega_{res} + \delta \omega \leq \omega \leq \omega_+$. However the two pairs $E^\parallel_-(k, -1) \rightarrow E^\parallel_-(k, 0)$, $E^\parallel_+(k, 0) \rightarrow E^\parallel_+(k, 1)$ and $E^\parallel_-(k, -1) \rightarrow E^\parallel_-(k, 0)$, $E^\parallel_+(k, 0) \rightarrow E^\parallel_+(k, 1)$ contribute $\chi''_+(\omega)$ in the interval $\omega \leq \omega \leq \omega_{res} - \delta \omega$. Here

$$\delta \omega = \sqrt{J_c^2 + g^2 \mu_B^2 h^2_{st} - |J_c|},$$

$$\omega_+ = g\mu_B h \pm |J_c| \pm \sqrt{J_c^2 + g^2 \mu_B^2 h^2_{st}}. \tag{20}$$

Finitely we may write the following formula

$$\chi''_+(\omega, T) \propto \chi_-(\omega, T) + \chi_+(\omega, T), \quad \omega_{min} \leq \omega \leq \omega_{res} - \delta \omega,$$

$$\chi''_-(\omega, T) \propto \chi_-(\omega, T), \quad \omega_{res} + \delta \omega \leq \omega \leq \omega_{max}, \tag{21}$$

where $\omega_{min} = \max(0, \omega_-)$ and $\omega_{max} = \omega_+.$

We see that the obtained line shape may be interpreted as twin asymmetric peaks separated by the gap. However it seems to us more fine to interpret it as a single-resonance contour with the gap around the resonance. The range of the spectrum and the width of the gap are controlled by cutoff parameters $\omega_{max} - \omega_{min}$ and $2\delta \omega$.

For low magnetic fields $h, h_{st} \ll \frac{|J_c|}{g\mu_B}$ a good approximation will be $\omega_{min} = 0$, $\omega_{max} = 2|J_c|$ and $\delta \omega = \frac{g^2 \mu_B^2 h^2_{st}}{2|J_c|}$. In this case according to (10) $2|J_c| = \Delta E_{magn}$ where the latter is the width of the one-magnon zone. The parameter $\Delta E_{magn}$ does not appear in the formula (19) but according to the following approximate formulas

$$\delta \omega \simeq \frac{g^2 \mu_B^2 h^2_{st}}{\Delta E_{magn}}, \quad \omega_{max} \simeq \Delta E_{magn}. \tag{22}$$
it has the sense of a scale parameter. In contrast to "mystical" ones often introduced ad hoc in Quantum Field Theory this parameter appears dynamically and has the clear physical interpretation and manifests the high-energy (spatial magnon dynamic) level at the low-energy (polarization magnon dynamic) one.

For $\omega_{res} \ll \omega \ll kT$ the magnetic susceptibility (21) has the following asymptotic:

$$\chi''(\omega, T) \propto \frac{1}{kT\omega}.$$  \hfill (23)

For $\omega \to \omega_{res} \pm \delta \omega$ asymptotics are the following:

$$\chi''(\omega_{res} \pm (\delta \omega + \epsilon), T) \propto \frac{1}{kT}e^{-\frac{(\Delta E_{\text{magn}})^2}{2kT}}.$$  \hfill (24)

The similar asymptotics at $\omega \to \omega_{res} \pm \delta \omega$ has the Gauss function:

$$\chi''_{\text{Gauss}}(\omega_{res} \pm (\delta \omega + \epsilon)) \propto \frac{1}{kT}e^{-\frac{(\Delta E_{\text{magn}})^2}{4kT \Delta E_{\text{magn}}}}.$$  \hfill (25)

## 5 Dipole-dipole and Dzyaloshinskii-Moria interactions

Traditionally ESR is applied for measuring dipole-dipole and Dzyaloshinskii-Moria interactions. In this section we shall briefly discuss both of them restricting ourselves on the case when the condition (4) is satisfied.

Dipole-dipole interaction along rungs induce an appearance of the following term

$$H_n^{\text{rung-dip}} = D_{\perp}(S_{1,n}^z S_{2,n}^z + \frac{1}{4}I) = \frac{D_{\perp}}{2}(S_{1,n}^z + S_{2,n}^z)^2, \quad D_{\perp} > 0.$$  \hfill (26)

In the absence of SMF the Hamiltonian (1)-(3), (25) is translational invariant and a one-magnon state has the form:

$$|1, k; \nu\rangle_{\text{magn}} = \sum_{n=-\infty}^{\infty} \sum_{j=-1,0,1} \zeta_j(k, \nu)e^{ikn}|1, n\rangle_j,$$  \hfill (27)

where the amplitudes $\zeta_j(k, \nu)$ satisfy the following Shrödinger equation:

$$(J(k) - g\mu_B \hbar \vec{S} + \frac{D_{\perp}}{2}\vec{S}^2)\zeta(k, \nu) = E(k, \nu)\zeta(k, \nu).$$  \hfill (28)

Here $J(k) = J_\perp - \frac{3}{2}J_c + J_c \cos k$. The cubic equation corresponding to the spectral problem (27) may be solved by the same method as the Eq. (14). Effect of the rung-dipole interaction reveals in splitting of resonance lines without any broadening.
The following terms

$$H_{n,n+1}^{\text{leg-dip}} = D_{ll} (S_{1,n}^z + S_{2,n}^z)(S_{1,n+1}^z + S_{2,n+1}^z), \quad (28)$$

$$H_{n,n+1}^{\text{DM}} = J_{DM} \cdot ([S_{1,n} \times S_{1,n+1}] + [S_{2,n} \times S_{2,n+1}] + [S_{1,n} \times S_{2,n+1}]
+ [S_{2,n} \times S_{1,n+1}]), \quad (29)$$

preserve the condition (4) and in some sense may be interpreted as dipole-dipole and Dzyaloshinskii-Morya interactions along legs and diagonals. However these terms do not contribute in the one magnon sector.

6 Conclusions

In the present paper we have obtained the exact formulas for one-magnon excitations in spin-ladders with exact singlet-rung ground state and staggered magnetic field oriented parallel or perpendicular to the constant magnetic field. In the parallel case we have calculated the one-magnon contribution to the ESR line shape. The latter is non-Lorentzian. It has the finite range and the gap around the resonance. Near the gap the line shape is Gaussian. For low magnetic fields both the width of the gap and the range of the spectrum are controlled by the the scale of the one-magnon zone. The presented results allow to conclude that the effects of staggered magnetic field are not perceptible in the spin-ladder material CaV$_2$O$_5$ [9].

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