Two-magnon Raman scattering in spin ladders with exact singlet-rung ground state

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Using coordinate Bethe ansatze we construct two-magnon states for the family of spin-ladder models with exact singlet-rung vacuum suggested by A. K. Kolezhuk and H.-J. Mikeska. The explicit formula for the zero-temperature Raman scattering cross section is derived. The corresponding line shapes are strongly asymmetric and their singularities originate only from bound states. This form of a line shape is in good correspondence to the experimental data.

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I. INTRODUCTION

Raman scattering in spin ladders was studied in a number of papers (see Refs. 1–9 and references therein). The obtained experimental data were analyzed by several theoretical approaches.[3,7,8] However, in none of these papers was the exact formula for the Raman cross section used. In the present paper we obtain the *exact* formula for the special class of spin-ladder models with exact singlet-rung vacuum. This family of models was first suggested in Ref. 10. The corresponding Hamiltonian $\mathcal{H}$ has the following form:

$$\mathcal{H}=\sum_{n=-\infty}^{\infty}H_{n,n+1}, \tag{1}$$

where

$$H_{n,n+1}=H^{stand}_{n,n+1}+H^{frust}_{n,n+1}+H^{\rm cyc}_{n,n+1}+H^{norm}_{n,n+1}, \tag{2}$$

and

$$H^{stand}_{n,n+1}=\frac{J_{\perp}}{2}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n}+\mathbf{S}_{1,n+1}\cdot\mathbf{S}_{2,n+1})+J_{\parallel}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{1,n+1}+\mathbf{S}_{2,n}\cdot\mathbf{S}_{2,n+1}), \tag{3}$$

$$H^{frust}_{n,n+1}=J_{frust}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n+1}+\mathbf{S}_{2,n}\cdot\mathbf{S}_{1,n+1}),$$

$$H^{cyc}_{n,n+1}=J_c((\mathbf{S}_{1,n}\cdot\mathbf{S}_{1,n+1})(\mathbf{S}_{2,n}\cdot\mathbf{S}_{2,n+1})+(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n})\times(\mathbf{S}_{1,n+1}\cdot\mathbf{S}_{2,n+1})-(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n+1})(\mathbf{S}_{2,n}\cdot\mathbf{S}_{1,n+1})),$$

$$H^{norm}_{n,n+1}=J_{norm}I.$$

Here $\mathbf{S}_{i,n}$ ($i=1, 2$; $n=-\infty,\ldots,\infty$) are spin-1/2 operators associated with cites of the ladder and I is an identity matrix. The auxiliary term $H^{norm}_{n,n+1}$ in (2) is needed only for normalization to zero of the lowest eigenvalue of the 16×16 matrix H of rung-rung interaction.

It was shown in Ref. 10 that when the following conditions,

$$J_{frust}=J_{\parallel}-\tfrac{1}{2}J_c,\quad J_{norm}=\tfrac{3}{4}J_{\perp}-\tfrac{9}{16}J_c, \tag{4}$$

$$J_{\perp}>2J_{\parallel},\quad J_{\perp}>\tfrac{5}{2}J_c,\quad J_{\perp}+J_{\parallel}>\tfrac{3}{4}J_c,$$

are satisfied, then the lowest (zero eigenvalue) eigenstate of H is $w\otimes w$, where w is the rung-singlet state. In this case the ground state of the Hamiltonian (1) has the simple tensor-product form:

$$|0\rangle=\prod_n\otimes w_n. \tag{5}$$

In order to obtain the full spectrum of H we shall also define the following triplet states:

$$f^k_n=(\mathbf{S}^k_{1,n}-\mathbf{S}^k_{2,n})w_n,\quad (\mathbf{S}^j_{1,n}+\mathbf{S}^j_{2,n})f^k_n=i\varepsilon_{jkm}f^m_n. \tag{6}$$

All other eigenstates of H are separated into the following sectors: singlet $f^k\otimes f^k$; triplet $\varepsilon_{ijk}f^j\otimes f^k$; quintet $t_{ijkl}f^j\otimes f^k$ with eigenvalues $\varepsilon_0=J_{\perp}-2J_{\parallel}$, $\varepsilon_1=J_{\perp}-J_{\parallel}-\frac{1}{4}J_c$, and $\varepsilon_2=J_{\perp}+J_{\parallel}-\frac{3}{4}J_c$; and two triplets $w\otimes f^k\pm f^k\otimes w$ with eigenvalues: $\varepsilon_{\pm}=(1/2)\big(J_{\perp}-\frac{3}{2}J_c\pm J_c\big)$. Here $t_{ijkl}=\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}-\frac{2}{3}\delta_{ij}\delta_{kl}$.

The Hamiltonian (1)–(3) commutes with the following magnon number operator $\mathcal{Q}=\Sigma_n Q_n$, where $Q_n=(3/4)I+\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n}$ is associated with the nth rung projection operator on triplet states.

II. THE TWO-MAGNON STATES

Corresponding to (1)–(4), one-magnon states were obtained in Ref. 10. Suggesting the following Bethe form for two-magnon states $|S,\beta\rangle$ (where S is the total spin and β the list of additional parameters),

$$|0;\beta\rangle=\sum_{m=-\infty}^{\infty}\sum_{n=m+1}^{\infty}a_0(m,n;\beta)\cdots w_{m-1}f^j_m w_{m+1}\cdots\times w_{n-1}f^j_n w_{n+1}\cdots, \tag{7}$$

$$|1;\beta\rangle_i=\sum_{m=-\infty}^{\infty}\sum_{n=m+1}^{\infty}a_1(m,n;\beta)\varepsilon_{ijk}\cdots w_{m-1}f^j_m w_{m+1}\cdots\times w_{n-1}f^k_n w_{n+1}\cdots, \tag{8}$$

$$|2;\beta\rangle_{ij}=\sum_{m=-\infty}^{\infty}\sum_{n=m+1}^{\infty}a_2(m,n;\beta)t_{ijkl}\cdots w_{m-1}f^k_m w_{m+1}\cdots\times w_{n-1}f^l_n w_{n+1}\cdots, \tag{9}$$

we obtain in the standard way[11] the following Schrödinger equation,

$$\frac{J_c}{2}[a_S(m-1,n;\beta)+a_S(m+1,n;\beta)+a_S(m,n-1;\beta)+a_S(m,n+1;\beta)]+(2J_\perp-3J_c)a_S(m,n;\beta)=Ea_S(m,n;\beta), \quad (10)$$

and the Bethe condition for amplitudes,

$$2\Delta_S a_S(n,n+1;\beta)=a_S(n,n;\beta)+a_S(n+1,n+1;\beta). \quad (11)$$

Here $\Delta_S=(\varepsilon_S-\varepsilon_+-\varepsilon_-)/(\varepsilon_+-\varepsilon_-)$.

For each S Eq. (11) has two solutions: the scattering solution,

$$a_S^{scatt}(m,n;k_1,k_2)=C_{S,12}\mathrm{e}^{i(k_1m+k_2n)}-C_{S,21}\mathrm{e}^{i(k_2m+k_1n)}, \quad (12)$$

with $C_{S,ab}=\cos(k_a+k_b)/2-\Delta_S\exp\left[i(k_a-k_b)/2\right]$, and the bound solution

$$a_S^{bound}(m,n;u)=\mathrm{e}^{iu(m+n)+v(m-n)}, \quad (13)$$

where the real parameters $v\geqslant 0$ and $-\pi<u\leqslant\pi$ satisfy the following condition:

$$\cos u=\Delta_S\,\mathrm{e}^{-v}. \quad (14)$$

From (14) and the non-negativity of v it follows that

$$|\cos u|\leqslant|\Delta_S|\leqslant\mathrm{e}^{v}. \quad (15)$$

The eigenvalues corresponding to (12) and (13) are

$$E_S^{scatt}(k_1,k_2)=2J_\perp-3J_c+J_c(\cos k_1+\cos k_2), \quad (16)$$

$$E_S^{bound}(u)=2J_\perp+(\Delta_S-3)J_c+\frac{J_c}{\Delta_S}\cos^2 u. \quad (17)$$

As we see from (12) and (13) the translation invariant states correspond to $a_S^{scatt}(m,n;k,-k)$, $a_S^{bound}(m,n;0)$, and $a_S^{bound}(m,n;\pi)$.

III. CALCULATION OF RAMAN CROSS SECTION

Following Sugai[2] we shall consider only the case when the incident and scattered light have parallel polarization directions, both lying in the plane of the ladder and forming an angle θ with respect to vertical bonds (rungs). The zero-temperature two-magnon Raman scattering cross section as a function of frequency and θ can be expressed using Fermi's golden rule:[3,4]

$$I(\omega,\theta)=\lim_{N\to\infty}\frac{2\pi}{2N+1}\sum_\alpha|\langle\alpha|\mathcal{H}^R(\theta)|0\rangle|^2\delta(\omega-E_\alpha), \quad (18)$$

where $2N+1$ is the number of rungs. Within the Fleury-Loudon-Elliot approach the effective Raman Hamiltonians $\mathcal{H}^R(\theta)$ have the following form[1,5] (we also take into account interactions across diagonals):

$$\mathcal{H}^R(\theta)=A_{\mathrm{leg}}\cos^2\theta\mathcal{H}^{leg}+A_{diag}[\cos^2(\theta+\gamma)\mathcal{H}^{d_1}+\cos^2(\theta-\gamma)\mathcal{H}^{d_2}]+A_{rung}\sin^2\theta\mathcal{H}^{rung}. \quad (19)$$

Here A_{leg}, A_{diag}, and A_{rung} are constants and γ is the angle between the rung and diagonal directions. Operators $\mathcal{H}^{rung}$, $\mathcal{H}^{leg}$, $\mathcal{H}^{d_1}$, and $\mathcal{H}^{d_2}$ are the following:

$$\mathcal{H}^{rung}=\sum_n\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n},\quad \mathcal{H}^{leg}=\sum_{i,n}\mathbf{S}_{i,n}\cdot\mathbf{S}_{i,n+1},\quad \mathcal{H}^{d_{1(2)}}=\sum_n\mathbf{S}_{1(2),n}\cdot\mathbf{S}_{2(1),n+1}. \quad (20)$$

Expressing $\mathcal{H}^{leg}$, $\mathcal{H}^{d_1}$, $\mathcal{H}^{d_2}$, and $\mathcal{H}^{rung}$ from the auxiliary operators,

$$\mathcal{H}^{\pm\pm}=\sum_n(\mathbf{S}_{1,n}\pm\mathbf{S}_{2,n})\cdot(\mathbf{S}_{1,n+1}\pm\mathbf{S}_{2,n+1}), \quad (21)$$

and taking into account Eq. (6), we represent $I(\omega,\theta)$ in the factorized form:

$$I(\omega,\theta)=\tfrac{1}{4}[(A_{leg}+A_{diag}\sin^2\gamma)\sin^2\theta+A_{diag}\cos^2\gamma\cos^2\theta]^2\times I_0(\omega), \quad (22)$$

where

$$I_0(\omega)=\lim_{N\to\infty}\frac{2\pi}{2N+1}\sum_\alpha|\langle\alpha|\mathcal{H}^{--}|0\rangle|^2\delta(\omega-E_\alpha). \quad (23)$$

Formula (22) expresses the polarization angle dependence of Raman cross section, however it may be applied in a straightforward way only for $\theta=m\pi/2$.[6]

From Eq. (6), translational and SU(2) invariance of $\mathcal{H}^{--}$ follows that only translation invariant singlet two-magnon states contribute to the formula (23). Separating the contributions from scattering and bound states we obtain:

$$I_0^{scatt}(\omega)=\lim_{N\to\infty}\sum_k\frac{\left|\sum_{n=-N}^{N}a(n,n+1;k,-k)\right|^2}{\sum_{n=-N+1}^{N}\sum_{m=-N}^{n-1}|a(m,n;k,-k)|^2}\times\delta(\omega-E_0^{scatt}(k,-k)), \quad (24)$$

$$I_0^{bound}(\omega)=\lim_{N\to\infty}\sum_{u=0,\pi}\frac{\left|\sum_{n=-N}^{N}a(n,n+1;u)\right|^2}{\sum_{n=-N+1}^{N}\sum_{m=-N}^{n-1}|a(m,n;u)|^2}\times\delta(\omega-E_0^{bound}(u)). \quad (25)$$

From (12) and (13) follows

$$\sum_{n=-N+1}^{N}\sum_{m=-N}^{n-1}|a_0^{scatt}(m,n;k,-k)|^2=4N^2(1-2\Delta_0\cos k+\Delta_0^2)+O(N), \quad (26)$$

$$\left|\sum_{n=-N}^{N}a^{scatt}(n,n+1;k,-k)\right|=4N\sin k+O(1), \quad (27)$$

$$\sum_{n=-N+1}^{N}\sum_{m=-N}^{n-1}|a_0^{bound}(m,n;u)|^2=\frac{2N}{e^{2v}-1}+o(N),\quad u=0,\pi, \tag{28}$$

$$\left|\sum_{n=-N}^{N}a^{bound}(n,n+1;u)\right|=(2N+1)e^{-v},\quad u=0,\pi. \tag{29}$$

Using the substitution $\Sigma_k \to [(2N+1)/2\pi]\int_0^{2\pi}dk$ we obtain from (26)–(29) the final expressions for the cross sections:

$$I_0^{scatt}(\omega)=\frac{4\Theta(1-x^2)\sqrt{1-x^2}}{J_c(1+\Delta_0^2-2x\Delta_0)}, \tag{30}$$

$$I_0^{bound}(\omega)=\frac{2\pi}{J_c}\left(1-\frac{1}{\Delta_0^2}\right)\Theta(\Delta_0^2-1)\,\delta\!\left(2x-\Delta_0-\frac{1}{\Delta_0}\right). \tag{31}$$

Here Θ is the step function and $x=(\omega-2J_\perp+3J_c)/2J_c$ is the rescaling parameter.

From (15) and (31) follows that the contributions from bound states $I_0^{bound}(\omega)$ exist only for $|\Delta_0|>1$. The behavior of I_0^{scatt} as a function of x also essentially depends on the parameter $\Delta_0=3/2-2J_\parallel/J_c$. When $\Delta_0=\pm1$ the formula (30) reduces and the line shape has a singularity at $x=\Delta_0$. For $\Delta_0=1$ it lies in the top of the two-magnon continuum, however for $\Delta_0=-1$ it is in the bottom. For $\Delta_0\neq\pm1$ the cross section I_0^{scatt} is a regular function of x and has the maximum at the point $x_{max}=2\Delta_0/\Delta_0^2+1$.

In order to study the line shape in more detail we shall find its inflection points. Calculating the second derivative of I_0^{scatt} with respect to x we obtain the following condition:

$$p(x,\Delta_0)=4\Delta_0(1+\Delta_0^2)x^3-12\Delta_0^2x^2-\Delta_0^4+6\Delta_0^2-1=0. \tag{32}$$

Since $p(\pm1,\Delta_0)=-(1\mp\Delta_0)^4$, the polynomial $p(x,\Delta_0)$ for $\Delta_0\neq\pm1$ has only 0 or 2 zeros in the interval $(-1,1)$. From standard calculation follows that $p(x,\Delta_0)$ has the maximum $p_{max}=-\Delta_0^4+6\Delta_0^2-1$ at the point $x=0$. It is evident now that for $p_{max}>0$ the line shape of I_0^{scatt} has two inflection points. From the straightforward calculation follows that $p_{max}>0$ only for

$$\Delta_-<|\Delta_0|<\Delta_+, \tag{33}$$

where $\Delta_\pm=\sqrt{3\pm2\sqrt{2}}$ ($\Delta_-\approx0.4142$, $\Delta_+\approx2.4142$). It may be easily proved in a straightforward way that $(\Delta_+-\Delta_-)^2=4$, so $\Delta_+-\Delta_-=2$.

In the case (33) the line shape near the x_{max} is similar to the van Hove singularity. For $\Delta_-<\Delta_0<\Delta_+$ this "singularity" lies near the top of the two-magnon continuum, however for $-\Delta_+<\Delta_0<-\Delta_-$ it is near the bottom. In both cases the line shape of the Raman scattering is strongly *asymmetric*. The case $p_{max}<0$ with no inflection points may be interpreted as a broad maximum. Some line shapes corresponding to different values of Δ_0 are presented in Fig. 1.

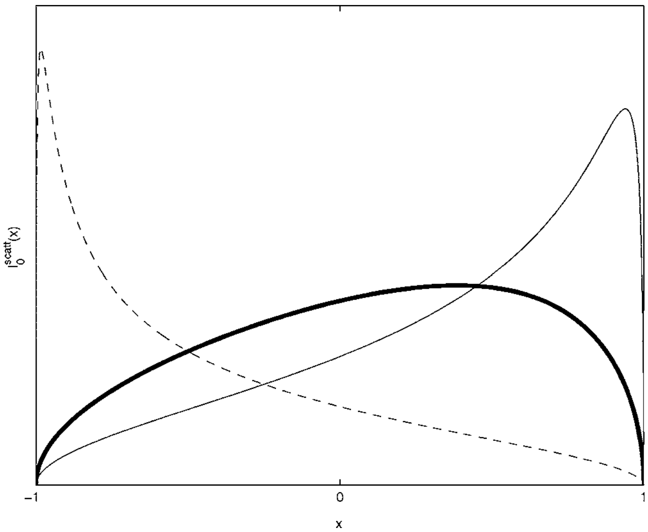

FIG. 1. The thick line: $\Delta_0=0.2$, the thin line: $\Delta_0=0.7$, and the dashed line: $\Delta_0=-1.2$.

As it follows from (16) and (17) for $\Delta_0\to\pm1+0^\pm$, the top (bottom) of the two-magnon continuum and the bound two-magnon state have the same energy: $2J\perp-3J_c\pm2J_c$. It was proposed in Ref. 3 that in this case the resonance between bound and scattering states leads to a redistribution of Raman intensity and merging of singularity. However, as we see from (30) and (31) in our model this conjecture fails. Moreover, the singularity in I_0^{scatt} appears only in the resonance $\Delta_0=\pm1$ cases.

IV. COMPARISON WITH EXPERIMENT AND DISCUSSION

Raman scattering in MgV_2O_5 and CaV_2O_5 were reported in Ref. 9. It was pointed that for both materials the corresponding line shapes are strongly asymmetric and have one maximum instead of two. This fact was considered as strange and some conjectures were suggested to interpret it. For example, it was supposed in Ref. 9 that in MgV_2O_5 there is no spin gap and the magnetic ordering is 2D, or the spin gap is so small (about 10 cm^{-1}) that it cannot be observed by the experimental resolution that was used. The asymmetry of the line shape for CaV_2O_5 was interpreted in Ref. 9 as originating from next-nearest neighbor interactions. In Ref. 7 it was conjectured that the second peak in the line shape of CaV_2O_5 is not observed because it is dominated by a phonon peak. In Ref. 3 it was conjectured that the asymmetry originates from resonance with a two-triplet bound state.

In our paper we have demonstrated that the line-shape asymmetry in spin-ladder Raman scattering is not something strange and outstanding, but may appear in a sufficiently big class of models. Of course we do not pretend that for some values of exchange parameters our toy model necessarily describes the real materials such as CaV_2O_5 or MgV_2O_5. Nevertheless perhaps the true ground state is in some sense "close" to our idealized one (5) and we may believe that our model correctly represents some general qualitative features of real materials. In this context we emphasize that the *exactly calculated* Raman scattering line shape may be strongly asymmetric without any additional assumptions such as next-nearest neighbor interactions, resonance with the bound state, or dominating phonon peak.

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