Raman cross section of spin ladders

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Abstract. We demonstrate that a two-triplet resonance strongly renormalizes the Raman spectrum of two-leg spin-ladders and moreover suggest this to be the origin of the asymmetry of the magnetic Raman continuum observed in CaV$_2$O$_5$.

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Magnetic Raman scattering is a powerful tool to investigate the total spin-zero excitations near zero momentum in low-dimensional quantum-spin systems [1]. In a recent Raman scattering study by Konstantinović and collaborators [2] a strongly asymmetric magnetic continuum, see fig. 1(b), has been observed in the spin-ladder compound CaV$_2$O$_5$. It has been realized by the authors of this study that the continuum defies an interpretation in terms of non-interacting two-triplet excitations as given in ref. [3]. The latter would imply two van-Hove-type intensity maxima, one at the lower and one at the upper edge of the two-triplet continuum. Noteworthy, the magnetic Raman intensity for the two-leg spin-ladder has been evaluated also by exact diagonalization (ED) [4]. Within the limits of finite system analysis the ED results are consistent with the observed intensity if the intra-rung coupling on the ladder is assumed to be strong in CaV$_2$O$_5$, moreover, the ED is incompatible with the non-interacting spectra of ref. [3]. While this clearly emphasizes the relevance of interaction effects, it is unfortunate that no simple physical picture can be extracted from the ED data to allow for a direct interpretation of the measured Raman spectrum.

In this brief note we clarify that the physical origin of the asymmetric Raman continuum of two-leg spin-ladders is a two-triplet bound state of total spin zero which merges with the two-triplet continuum at small wave vector to form a resonance. Our analysis is focussed on the limit of strong intra-rung coupling which is one likely scenario also for the magnetic properties of CaV$_2$O$_5$ [4]. In this limit we can profit from an exact evaluation of the two-triplet propagator which has been carried out including all two-triplet interactions in a different study of phonon-assisted two-triplet optical absorption (PTA) of spin-ladders [4].

The Hamiltonian of the two-leg spin-ladder reads

\[
H = \sum_{l,\alpha} [S_{l\alpha}^x S_{l+1\alpha}^x + \lambda (S_{l\alpha}^y S_{l+1\alpha}^y + S_{l\alpha}^z S_{l+1\alpha}^z)]
\]  

where \( S_{l\alpha}^\alpha \) with \( \alpha = x, y, z \) is a spin-1/2 operator on site \( l \) of leg \( \mu \) and \( H \) is measured in units of \( J_\perp \) with \( \lambda = J_\parallel /J_\perp \).

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where \( R \) depends on the polarizations of the incident and scattered light [3], \( H_R \) is similar to the vertex for PTA [3], simplified however by the lack of an additional summation over phonon coordinates. The Raman intensity \( I(\omega) \) at zero temperature is obtained from Fermi’s golden rule

\[
I(\omega) = 2\pi \sum_f |\langle f|H_0|0\rangle|^2 \delta(\omega - E_f)
\]

\[
= -2 \text{Im} \sum_{q,q'} |\langle 0|H_R^\dagger|q\rangle| \frac{1}{z - H|q\rangle \langle q'|H_R|0\rangle}
\]

where \( z = \omega + i0^+ \). \( |0\rangle \) \((|f\rangle\) are the interacting ground (excited) states with energy \( 0 \) \((E_f\) and total momentum and spin zero. For \( \lambda \ll 1 \) and following [3] \( |0\rangle \) is a product of rung-singlets and \( |f\rangle \) are interacting two-triplet excitations. Neglecting quantum fluctuations which change the number of triplets only at \( O(\lambda^2) \) the states \( |f\rangle \) can be expanded in terms of an appropriately symmetrized basis \(|q\rangle\) of two-triplet rung excitations

\[
|q\rangle = \frac{1}{\sqrt{N(N-1)}} \sum_{l,m} \text{sgn}(l - m) \sin(q(l - m)) |lm\rangle
\]

where \(|lm\rangle = \sum_\alpha |t_{l\alpha}t_{m\alpha}\rangle/\sqrt{3} \) refers to a singlet combination of two rung triplets created within \(|0\rangle \). The states \(|q\rangle\) resemble all spin-zero two-triplet plane-waves of zero total momentum constrained by the symmetry \(|t_{l\alpha}t_{m\beta}\rangle = |t_{m\beta}t_{l\alpha}\rangle \) and the hard-core condition \(|t_{l\alpha}t_{l\beta}\rangle = 0 \). The remaining resolvent in (3) can be evaluated in closed form.

\[
|q\rangle = \frac{1}{\sqrt{N(N-1)}} \sum_{l,m} \text{sgn}(l - m) \sin(q(l - m)) |lm\rangle
\]
by a T-matrix resummation (see 6)

\[ \lambda I(\omega) = \frac{3}{4} \text{ln} \left( \sqrt{1 - \frac{4}{2 + \omega}} - 1 \right) \]  

In the limit of \( \lambda \ll 1 \), the intensity is a function of the rescaled Raman-shift \( \tilde{\omega} = (\omega + i0^+ - 2)/\lambda \) only. While the largest energy scale, i.e., the two-triplet hard-core, is incorporated in the states \( |q\rangle \) by construction, the T-matrix resummation accounts for both, the dispersion and the nearest-neighbor (NN) attraction which is mediated on the two-particle level by Hamiltonian \( \mathbf{H} \).
The thick solid line in fig. 1(b) is consistent with the intensity distribution obtained from ED \( \mathbf{H} \).

The dotted line in 1(a) depicts the bare Raman intensity \( \mathbf{H} \) which results from neglecting the two-triplet on-site hard-core as well as the NN-attraction. Displaying two van-Hove singularities this spectrum fails to explain the observed magnetic line-shape.

The physical origin of the asymmetric line-shape is clarified in the inset of fig. 1(a). While Raman scattering detects only zero momentum excitations the inset reproduces the interacting two-triplet spectrum in the spin-zero channel for \( \lambda \ll 1 \) over all of the Brillouin zone from ref. \( \mathbf{H} \). Apart from the bare two-triplet continuum this spectrum shows a bound-state induced by the two-triplet interactions which merges with the continuum at zero momentum. This leads to a resonance at the bottom of the continuum and to the asymmetric redistribution of the Raman intensity. This resonance feature has to be contrasted against Raman intensities in other low-dimensional quantum spin systems where bound states tend to occur as sharp excitations within the spin gap \( \mathbf{H} \).

Finally, based on the results of high-order series expansion \( \mathbf{H} \) it is tempting to speculate on the evolution of the Raman continuum as \( \lambda \rightarrow 1 \). In that limit the spin-zero bound-state merges with the continuum already at finite momentum. Therefore, as \( \lambda \) increases one might expect the resonance to shift further into the center of the continuum. This suggests that an analysis analogous to this work of Raman data on compounds containing spin-ladders with \( \lambda \sim 1 \), e.g., \((\text{Ca,La})_4\text{Cu}_2\text{O}_{11}\), should be interesting to perform.

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