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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_2O_5.

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Rapid Note

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 Figure 1b, has been observed in the spin-ladder compound CaV_2O_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 reference [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 limitations 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_2O_5, moreover, the ED is incompatible with the non-interacting spectra of reference [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_2O_5 [2,5]. 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 [6].

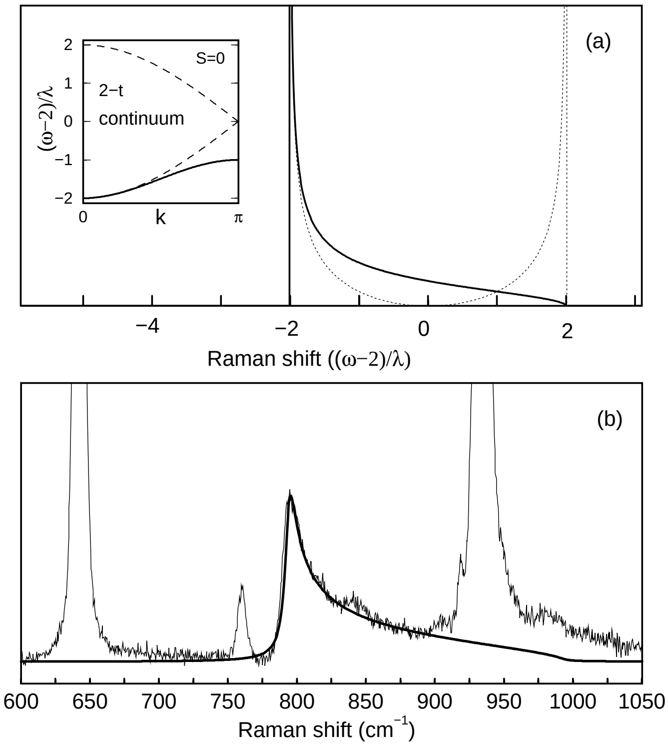

Fig. 1. (a) Solid line: Raman intensity (6). Dotted line: bare Raman intensity neglecting two-triplet interactions. Inset: $S = 0$ two-triplet spectrum of spin ladders. (b) Thin solid line: experimental Raman spectrum (after [2]). Thick solid line: fit of theory to experiment, see text.

The Hamiltonian of the two-leg spin-ladder reads

$$H = \sum_{l,\alpha} [S^\alpha_{1l} S^\alpha_{2l} + \lambda (S^\alpha_{1l} S^\alpha_{1l+1} + S^\alpha_{2l} S^\alpha_{2l+1})] \qquad (1)$$

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

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Magnetic light scattering is described by the Loudon–Fleury vertex [7], which for the two-leg spin-ladder is

$$H_R = R \sum_{l,\alpha} (S^\alpha_{1l} S^\alpha_{1l+1} + S^\alpha_{2l} S^\alpha_{2l+1}) \quad (2)$$

where R comprises of a materials parameter, the amplitudes of the incident and scattered light, and a form factor describing the polarization geometry. We refrain from a discussion of the polarization dependence which leads to a trivial rescaling of the Raman intensity [8]. H_R is formally identical to the vertex for PTA in equation (7) of [6] by combining all terms in the latter equation involving the effective charges, phonon coordinates, and electric field into the single constant R. This allows for a direct application of the formalism of [6]. The Raman intensity $I(\omega)$ at zero temperature is obtained from Fermi's golden rule

$$I(\omega) = 2\pi \sum_f |\langle f|H_R|0\rangle|^2 \delta(\omega - E_f) \quad (3)$$

$$= -2 \ \mathrm{Im} \sum_{q,q'} [\langle 0|H_R^\dagger|q\rangle\langle q|\frac{1}{z-H}|q'\rangle\langle q'|H_R|0\rangle] \quad (4)$$

where $z = \omega + \mathrm{i}0^+$. $|0\rangle$ ($|f\rangle$) are the interacting ground (excited) states with energy 0 (E_f) and total momentum and spin zero. For $\lambda \ll 1$ and following [6] $|0\rangle$ 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

$$|q\rangle = \frac{1}{\sqrt{N(N-1)}} \sum_{l,m} \mathrm{sgn}(l-m) \mathrm{sin}(q(l-m))|lm\rangle \quad (5)$$

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 (4) can be evaluated in closed form by a T-matrix resummation (see [6])

$$\frac{\lambda I(\omega)}{R^2} = \frac{3}{4} \mathrm{Im} \left(\sqrt{1 - \frac{4}{2+\tilde{\omega}}} - 1 \right). \quad (6)$$

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 (1). Discarding an overall prefactor (6) stems directly from equations (14, 15) of [6] by replacing p_k with R, by omitting the sum on the momentum k of the phonon which is relevant only in reference [6], and by setting $k = 0$ instead. Using a different technique the $S = 0$ two-triplet dynamics has also been investigated to leading order in λ by Uhrig and Schulz [9] with a spectrum identical to our findings. This further corroborates our results on the Raman intensity.

The thick solid line in Figure 1a is the intensity (6). The thin solid line in Figure 1b is the intensity measured on CaV_2O_5 at 10 K (reproduced from [2]). In addition to phonons the observed spectrum shows a strongly asymmetric line with an onset at 795 cm^{-1}. The thick solid line in Figure 1b is a comparison of (6) to the experiment which results (i) from using $\lambda = 0.11$ which is motivated by the ratio of the continuum line-width to the Raman-gap and which is consistent with [2], where $\lambda = 0.1$ was suggested, (ii) by setting $J_\perp = 447$ $\mathrm{cm}^{-1} \equiv 643$ K which agrees with [2] and is needed to obtain a continuum onset at 795 cm^{-1} for $\lambda = 0.11$, (iii) by including a broadening of $\sim$3 cm^{-1} by setting $\omega + \mathrm{i}0^+ \rightarrow \omega + \mathrm{i}0.007$ to account for instrumental resolution, and (iv) by adjusting the arbitrary y-axis scales for a reasonable match of the absolute intensities. We note that in [5] a slightly different set of parameters, *i.e.*, $J_\perp = 667(3)$ K and $\lambda = 0.1$–0.16 has been extracted from mean-field and Quantum-Monte-Carlo analysis of thermodynamic bulk data. The latter analysis incorporates effects of inter-ladder couplings. Quantitative agreement of the type displayed in Figure 1b does not result from the parameter set of [5], yet a qualitative consistency remains. While more ambitious fitting procedures can be envisaged the preceding is sufficient to claim that the agreement between experiment and our theory is very good. Moreover, we note that the solid line in Figure 1b is consistent with the intensity distribution obtained from ED [4].

The dotted line in Figures 1a depicts the bare Raman intensity [3] 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 Figure 1a. 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 reference [6]. 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 [1]. It is important to realize that both, the hard-core repulsion inherent in the two-triplet wave-function (5) and the NN triplet-attraction have two very distinct effects on the final shape of the Raman spectrum. While the hard-core repulsion completely suppresses *both* van-Hove singularities observed in the bare Raman spectrum [10] it is only due to the NN-attraction that the resonance peak appears at the lower edge of the fully interacting Raman continuum.

Finally, based on the results of high-order series expansion [11] it is tempting to speculate on the evolution of the Raman continuum as $\lambda \to 1$. In that limit the spin-zero bound-state merges with the continuum already at finite momentum. Therefore, as λ 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.* $(Ca,La)_{14}Cu_{24}O_{41}$, should be interesting to perform.

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