Raman Scattering cross section of Spin Ladders

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The Raman scattering spectra from magnetic excitations in an antiferromagnetic spin-1/2 two leg ladder is investigated for weak and strong interladder coupling. In the first case, a cusp in the Raman intensity is obtained at a frequency twice the gap. In the second case, a peak at twice the gap replaces the cusp. We discuss the relevance of our calculation to recent experiments on CuV\textsubscript{2}O\textsubscript{5} and Sr\textsubscript{14}Cu\textsubscript{24}O\textsubscript{41}.

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Raman scattering is an experimental technique that has provided valuable informations about the spin dynamics in quasi-one dimensional antiferromagnets in the recent years. Since Raman scattering is sensitive to singlet excitations, this technique is complementary of neutron diffraction which is sensitive to triplet excitations. It has been used to probe spin 1/2 chains, spin 1 chains, spin Peierls systems, and spin ladders. In particular, Raman scattering has been very useful in the analysis of magnetic excitations in the spin-Peierls compound CuGeO\textsubscript{3}. The bosonized theory of dimerized spin 1/2 states is believed to describe the dimerized low temperature phase of spin-Peierls systems predicts the appearance of a singlet bound state of two triplet excitations at an energy $\sqrt{3}\Delta$, where $\Delta$ is the spin gap\textsuperscript{[1]}\textsuperscript{[2]}. Such singlet bound state has been successfully observed in Raman scattering experiments on CuGeO\textsubscript{3} for $T < T_{SP}$ at an energy 1.79$\Delta$, close to the theoretical prediction. Moreover, the peak was not observed in the uniform phase($T > T_{SP}$), showing that it is characteristic of the dimerized phase\textsuperscript{[1]}. For $T > T_{SP}$, a broad band of magnetic excitations is observed\textsuperscript{[3]}. The theoretical analysis of magnetic Raman scattering is based on the Fleury-Loudon Hamiltonian\textsuperscript{[4][5]}\textsuperscript{[6]}, that describes the interaction of photons with magnetic excitations. There exists at present a certain amount of litterature on the theory of Raman scattering from dimerized spin chains, both analytically\textsuperscript{[7][8]}\textsuperscript{[9]} and numerically\textsuperscript{[10]}. The case of frustrated spin chains has also been investigated\textsuperscript{[11]} in relation with the Raman spectra of CuGeO\textsubscript{3} at $T > T_{SP}$. The theory of Ref.\textsuperscript{[12]} reproduces well the features of the spectrum at $T > T_{SP}$\textsuperscript{[13]}\textsuperscript{[14]}. The application of Raman scattering to probe the singlet excitations of two leg ladders is more recent\textsuperscript{[15]}. In spin ladder systems, magnetic peaks in the Raman intensity were observed at twice the spin gap. From the theoretical point of view, some numerical calculations are available\textsuperscript{[16]} but no analytic expression of the Raman intensity has been derived so far. In order to fill this gap, we discuss in the present work the Raman spectrum of an antiferromagnetic spin-1/2 ladder. After recalling some basic results on the Fleury-Loudon theory of magnetic Raman scattering, we will consider first the Majorana fermion approach valid for weak coupling and then the Bond Operator Technique (BOT) valid for the strong coupling case. The Majorana fermions approach leads to a cusp in the Raman intensity at twice the gap, in disagreement with experiment. We discuss briefly what could be missing in the Majorana fermions description. On the other hand, the BOT predicts correctly the presence of peaks in the Raman intensity at twice the gap.

We consider two coupled antiferromagnetic $S = 1/2$ Heisenberg chains, whose Hamiltonian is

$$H = J \sum_i \left( \vec{S}_{1,i} \cdot \vec{S}_{1,i+1} + \vec{S}_{2,i} \cdot \vec{S}_{2,i+1} \right) + J_\perp \sum_i \vec{S}_{1,i} \cdot \vec{S}_{2,i}$$

(1)

where $J > 0$ and $J_\perp > 0$ denotes the intra- and inter-chain antiferromagnetic interactions, respectively. The interaction of light with the antiferromagnetic fluctuations is described by Loudon-Fleury's photon-induced super-exchange operator

$$H_R = \sum_{i,j} \left( \vec{E}_{i} \cdot \vec{S}_{ij} \cdot \vec{E}_{j} \cdot \vec{S}_{ij} \right)$$

(2)

where $\vec{E}_i$ is the incident (scattered) electric field, and $\vec{S}_{ij}$ is a unit vector connecting the sites $i$ and $j$, at which the spins $\vec{S}_i$ and $\vec{S}_j$ are located. A derivation of formula\textsuperscript{[13]} starting from the Hubbard Hamiltonian can be found in Ref.\textsuperscript{[16]}

The Raman cross section\textsuperscript{[17]}\textsuperscript{[18]} can be expressed as a function of the retarded Raman response function as:

$$\frac{d^2 \sigma}{d\Omega d\omega} = \frac{\omega_1 \omega_2}{2\pi c^4 V} \frac{n_2}{n_1} \frac{1}{1 - e^{-\beta \hbar \omega}} \text{Im} \chi_R(\omega)$$

(3)
ω₁ and ω₂ are the frequencies of the incoming and scattered radiation, respectively, ω = ω₂ − ω₁, n₁ and n₂ are the respective refractive index. V is the volume of the crystal and c the velocity of light. The retarded linear response function χ_R(ω) is defined as:

\[ \chi_R^{ren}(ω) = \frac{i}{\hbar} \int_0^\infty e^{i(ω+0) t} \text{Tr} \left\{ \sqrt{Z}^{-1} e^{-βH} [H_R(t), H_R(0)] \right\}, \]

where \( Z = \text{Tr} e^{-βH} \) and \( H_R \) is the Loudon-Fleury Hamiltonian \([3]\).

By inserting the resolution of identity in \((4)\), the Raman intensity can be written as

\[ \frac{d^2 \sigma}{d\Omega dω_2} \propto \frac{1}{\hbar} \sum_{n,m} e^{-βE_n} |⟨\Psi_n | H_R | \Psi_m⟩|^2 \times \delta(ω - (E_n - E_m)/\hbar), \]

where \( |\Psi_{n(m)}⟩ \) are eigenstates with energies \( E_{n(m)} \). Such formula can be easily interpreted as a Fermi golden rule averaged over the Boltzmann weight. To get informations on two-magnons scattering processes we should perform a symmetry analysis of the matrix elements appearing in \((3)\), and discuss selection rules. Since the spin ladder Hamiltonian is invariant under translation along the legs, SU(2) rotation, and mirror along the leg direction, an eigenstate should be characterized by a (lattice) momentum defined modulo 2π/a (where \( a \) is the lattice spacing), a spin and its parity under leg exchange. The Raman operator defined in \((3)\) is rotationally and translationally invariant, and still invariant under leg exchange. As a result, the selection rules impose that the states \( |\Psi_n⟩ \) and \( |\Psi_m⟩ \) have the same spin, momentum and parity under leg exchange. This implies, in particular, that at \( T = 0 \), transitions will only take place to states of total momentum zero, spin zero and same parity as the ground state. Let us now turn to concrete calculations. We consider the scattering for \( E_1 \) and \( E_2 \), parallel to the rung direction, thus we have

\[ H_R = \frac{\text{cste}}{2} E_1 E_2 \sum_i \delta_i \delta_{2,i}. \]

In the following, we will evaluate the Raman intensity in the weak coupling and in the strong coupling limit using the standard Matsubara technique to calculate the correlator \( \chi_R(ω) \).

To evaluate the time ordered Raman susceptibility for the weakly coupled chains, we will employ the Majorana fermion representation of the spin-ladder Hamiltonian \([4]\) introduced by Shelton, Nersesyan and Tsvelik in Ref. \([23]\). The effective Hamiltonian is expressed in terms of four interacting Majorana fermions. They comprise a degenerate triplet \( \xi^\nu(x) \) (\( \nu = \text{Left, Right} \)) with bare mass \( m_1 = m = J_L \) and a singlet, \( \rho_0(x) \) with bare mass \( m_s = -3m \). It has been argued in Ref. \([23]\) that the effect of interactions was merely to renormalize the bare masses, so that interactions could be neglected. With this approximation, the spin ladder is described by the following effective Hamiltonian:

\[ H = \sum_{a=1,2,3} H_m[\xi^a] + H_{-3m}[\rho], \]

where

\[ H_m[\chi] = \left\{ -i \frac{v}{2} \left[ \chi_R \partial_x \chi_R - \chi_L \partial_x \chi_L \right] - i \mu \chi_L \chi_R \right\}, \]

where \( \mu \) stands for the triplet or singlet mass, and \( \chi \) is the corresponding triplet or singlet operator. The thermal Green’s function for the left and right moving triplet and singlet Majorana fermions are defined by:

\[ G^\chi_{\mu}(k,iω_n) = \left\langle \frac{\xi^\mu_{\alpha}(−ω_n, k)\xi^\alpha_{\mu}^{\dagger}(ω_n, k)}{2} \right\rangle, \]

\[ G^\alpha_{\mu}(k,iω_n) = \left\langle \frac{\rho_\mu(−ω_n, k)\rho_\mu^{\dagger}(ω_n, k)}{2} \right\rangle, \]

whose explicit expressions are

\[ G^α_{RR}(k,iω_n) = G^α_{LL}(−k,iω_n) = -\frac{iω_n + νk}{ω_n^2 + ν^2k^2 + m_s^2} \]

\[ G^α_{RL}(k,iω_n) = G^α_{LR}(−k,iω_n)^* = -\frac{im_s}{ω_n^2 + ν^2k^2 + m_s^2} \]

where \( α \) stands for t (triplet) or s (singlet), and \( ω_n = (2n + 1)π/β \) are the fermion Matsubara frequencies. In terms of Majorana fermions, the Raman operator \( \gamma \sum_i \vec{S}_i \cdot \vec{S}_{2,i} \), with \( γ \) a constant, is expressed by

\[ H_R = γ_1 ℓ_R ℓ_L + γ_2 ρ R ρ L, \]

where \( γ_1 = m_t \gamma_1 \) and \( γ_2 = m_s \gamma_2 \). To arrive at this expression, the marginal term, already neglected in the derivation of the Hamiltonian \([3]\), has been discarded. Injecting this expression into the definition of the Raman susceptibility \((3)\) and applying Wick’s theorem, the time ordered expectation value at finite temperature can be written as:

\[ \chi_R(−ω_n) = \frac{1}{β} \sum_{ν_{a,α}} γ_α^2 \int \frac{dq}{2π} G^α_{RL}(q,iω_n)G^α_{LR}(−q,i(ω_n − ν_ν)) \]

\[ -G^α_{RR}(−q,iω_n)G^α_{LL}(−q,i(ω_n − ν_ν)). \]

Explicitly, we have to compute the following integral and sum over the Matsubara frequencies:

\[ \chi_R(−ω_n) = \frac{1}{β} \sum_{ν_{a,α}} γ_α^2 \times \int \frac{dq}{2π} \left\{ \frac{m_α^2 - (ivq_q + ν_ν)}{(ν_ν^2 + (vq)^2 + m_α^2)((ω_n − ν_ν)^2 + (vq)^2 + m_α^2)} \right\}. \]

In order to evaluate the Matsubara sum in \((12)\), we have to determine the residues of the four poles of the expression \((12)\) and multiply every residue with the value of the Fermi function \( n_F(z) = 1/(\exp(βz) + 1) \) at the pole. Adding the four terms together yields
\[ \chi_R(i\omega_n) = -\sum_\alpha \gamma_\alpha^2 \int \frac{dq}{\varepsilon_\alpha(q)} (1 - 2n_F(\varepsilon_\alpha(q))) \times \]
\[ 2\nu \left[ \frac{i\omega_n + 2\nu q}{\omega_n^2 + 4\varepsilon_\alpha(q)^2} \right], \tag{13} \]

where we have introduced the notation \( \varepsilon_\alpha(q) = \sqrt{\nu^2 + m_\alpha^2} \). Thus performing the analytic continuation (\( i\omega_n \to \omega + i0 \), taking the imaginary part and performing the integral over \( q \), we finally get:

\[ \text{Im}\chi_R(\omega) = \pi \sum_\alpha \text{tanh} \left( \frac{\omega}{4k_BT} \right) \gamma_\alpha^2 \frac{\sqrt{\omega^2 - 4m_\alpha^2}}{2\omega} \]
\[ \times \Theta(|\omega| - 2m_\alpha) \tag{14} \]

Formula (14) implies the existence of a cusp singularity in the Raman intensity at twice the spin gap due to the triplet excitations and another singularity at six times the spin gap due to the singlet modes. As result, the noninteracting Majorana fermions representation does not reproduce the Raman peak experimentally observed. The spectra predicted by (14) is plotted on figure 3. The absence of signal for \( \omega \) smaller than twice the gap is in qualitative agreement with numerical simulations. It would be interesting to determine whether treating properly the interactions between the Majorana Fermions can reproduce the experimental peak at twice the spin gap. We now turn to a strong-coupling analysis of the Raman susceptibility, using the bond operator representation\(^{24}\) of quantum S=1/2 spins used by Gopalakrishnan, Rice and Sigrist\(^{25}\) in their mean field approach to spin ladders. In this representation, one starts from weakly coupled rungs and introduces on each rung a singlet \( s^\dagger \) and three triplets \( t_{i,\alpha}^\dagger (\alpha = x, y, z) \) boson creation operators, that span the Hilbert space of a single rung when acting on a vacuum state. Since the rung can be in either the singlet or one of the triplet states, the condition:

\[ s^\dagger s + \sum_\alpha t_{i,\alpha}^\dagger t_{i,\alpha} = 1 \tag{15} \]

has to be satisfied by the physical states. The representation of the spins \( S_1 \) and \( S_2 \) in terms of these singlet and triplet operators, is derived in Ref.\(^{24}\). Substituting this operator representation of spins into the original Hamiltonian, one ends up with an Hamiltonian quartic in boson fields. Treating the singlet operator in a mean field approximation and neglecting interactions among the triplets, one obtains the following Hamiltonian quadratic in triplet operators:

\[ H_{\text{MF}} = (J_z/4 - \mu) \sum_{i,\alpha} t_{i,\alpha}^\dagger t_{i,\alpha} + \frac{J_z^2}{2} \sum_{i,\alpha} (t_{i,\alpha}^\dagger + t_{i,\alpha}) \langle t_{i+1,\alpha}^\dagger (t_{i+1,\alpha}^\dagger + t_{i+1,\alpha}). \tag{16} \]

The chemical potential term \( \mu \) guarantees that the condition (15) is satisfied on average. This Hamiltonian can be solved by Green’s method function. One, first, introduces the four Green’s functions \( G_{i,\alpha}(\tau) = -\langle T_{\tau} t_{i,\alpha}(\tau) t_{0,\alpha}^\dagger(0) \rangle \), \( \tilde{G}_{i,\alpha}(\tau) = -\langle T_{\tau} t_{i,\alpha}(\tau) t_{0,\alpha}(0) \rangle \), \( F_{i,\alpha}(\tau) = -\langle T_{\tau} t_{i,\alpha}(\tau) t_{0,\alpha}^\dagger(0) \rangle \) and their Fourier transforms. We have:

\[ G(k, i\omega_n) = -\left[ \tilde{G}(k, i\omega_n) \right]^\dagger = \frac{i\omega_n + \Delta_k}{\omega_n^2 + \omega^2} \tag{17} \]
\[ F(k, i\omega_n) = F^\dagger(k, i\omega_n) = \frac{2\Delta_k}{\omega_n^2 + \omega^2} \]

where \( \nu_n = \frac{2m_\alpha^2}{\nu^2} \) and the following notation has been introduced: \( \omega_3^2 = \Delta_k^2 - (2\Delta_k)^2 \), with \( \Delta_k = J_z/4 - \mu - J_z^2 S^2 \cos k \) and \( \Delta_k = J_z/4 - \mu + J_z^2 S^2 \cos k \), recovering the dispersion relation predicted by Gopalakrishnan, Rice and Sigrist\(^{25}\). As shown in Ref\(^{26}\), the parameters \( \mu \) and \( s \) are determined by solving the self-consistent saddle point equations. Let us now turn to the calculation of the Raman intensity.

The Raman intensity is proportional to \( \text{Im}\chi_R(i\omega_n \to \omega + i0) \) where:

\[ \chi_R(i\omega_n) = \sum_{\alpha,\beta} \int_0^\beta d\tau \tilde{c}_{\omega_n} \langle T_{\tau} (t_{\alpha}^\dagger t_{\alpha})(\tau)(t_{\beta}^\dagger t_{\beta})(0) \rangle \tag{18} \]

By using the definition (18) and applying Wick’s theorem, the following expression for the Raman susceptibility is obtained:

\[ \chi_R(i\omega_n) = \beta^{-1} \sum_{\nu_n} \int \frac{dk}{2\pi} \left\{ G(k, i\nu_n) G(k, i\nu_n - i\omega_n) + F(k, i\nu_n) F^\dagger(k, i\nu_n - i\nu_n) \right\}. \tag{19} \]

Performing the usual linear response calculation\(^{22}\), we obtain as a final result:

\[ \text{Im}\chi_R(\omega) = \frac{\coth \left( \frac{\omega}{4k_BT} \right) \left( \frac{\omega}{2(J_z/4 - \mu)} \right)^2 - 1}{4\omega \left( \frac{2J_z^2}{J_z/4 - \mu} \right)^2 - 1^2} \tag{20} \]

The Raman scattering spectra will show two peaks, one at energy \( \omega = 2\omega_\tau = 2\Delta_s \) corresponding to the bottom of the triplet band, and a second one at \( \omega = 2\omega_0 \), corresponding to the top of the triplet band. Close to the critical frequency \( \omega^* \), \( I(\omega) \sim (\omega - \omega^*)^{-1/2} \) this behavior can be easily understood by a density of states argument. The resulting spectra is plotted in figure 3. No signal is obtained for \( \omega < 2\Delta_s \) in agreement with numerics. Let us note that in recent experiments, a Raman scattering peak at twice the gap is observed in CaV\textsubscript{2}O\textsubscript{4}\text{Cl} where the spin-gap and the exchange constant are estimated to be \( \Delta_s \sim 400\text{cm}^{-1} \) and \( J_\perp \sim 640\text{K} \). These results are in qualitative agreement with our theory. In the case of
Sr$_2$Cu$_2$O$_{4.1}$, the situation is more complicated due to the coexistence in the structure of dimerized spin chains, having a spin gap $\Delta_{chain} = 12$meV and of spin ladders having a spin gap $\Delta_{ladder} \approx 32$meV. In Ref. 4, a peak was observed at 570cm$^{-1}$ $\approx 71$meV in Raman scattering experiments on polycrystalline samples. According to our theory, this would lead to a spin gap of $\approx 35$meV, in agreement with neutron scattering datas.

A more recent investigation or Raman scattering on single crystals identifies a peak at 498cm$^{-1}$ as the Raman peak associated with the gap. The peak at 569cm$^{-1}$ is identified with a (0,0) gap. According to the authors of Ref. 4, the other peaks are associated with bound states or single magnon light scattering. It is known that bound states of magnetic excitations can be formed below the gap in a spin ladder. In our treatment, we have been neglecting them altogether. They should give rise to peaks below the threshold 2$\Delta$, as has been observed in experiments. This problem is under investigation.

To summarize, we have considered Raman scattering in a spin ladder both in the weak coupling and the strong coupling approximation. We have shown that only the strong coupling treatment gave rise to peaks in the Raman intensity. Future directions include the consideration of the effect of bound states on the Raman spectra. We thank T. Giamarchi and O. Parcollet for their remarks on the manuscript. E. O. acknowledges discussion with K. Damle on the bound states in a spin ladder. E. O. acknowledges support from NSF under grant DMR 96-14999.

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**FIG. 1.** Raman intensity in arbitrary units for $J_\perp \ll J$ at $T = 0K$ obtained from the Majorana fermion approach. The frequency $\omega$ is measured in units of the gap.

**FIG. 2.** Raman intensity in arbitrary units for $J_\perp/J = 2$ at $T = 0K$. The frequency $\omega$ is measured in units of the gap.