Raman scattering cross section of spin ladders

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The Raman scattering spectra from magnetic excitations in an antiferromagnetic spin-$\frac{1}{2}$ 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 CaV_2O_5 and $Sr_{14}Cu_{24}O_{41}$.

Raman scattering is an experimental technique that has provided valuable information 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,[1] spin-1 chains,[2] spin-Peierls systems,[3–6] and spin ladders.[7–9] In particular, Raman scattering has been very useful in the analysis of magnetic excitations in the spin-Peierls compound $CuGeO_3$. The bosonized theory of dimerized spin-1/2 states that 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 Δ is the spin gap.[10] Such a singlet bound state has been successfully observed in Raman scattering experiments on $CuGeO_3$ for $T<T_{SP}$ at an energy 1.79Δ, 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.[4,11] For $T>T_{SP}$, a broad band of magnetic excitations is observed.[4] The theoretical analysis of magnetic Raman scattering is based on the Fleury-Loudon Hamiltonian,[12,13] that describes the interaction of photons with magnetic excitations. There exists at present a certain amount of literature on the theory of Raman scattering from dimerized spin chains, both analytical[10,14,15] and numerical.[16] The case of frustrated spin chains has also been investigated,[17] in relation with the Raman spectra of $CuGeO_3$ at $T>T_{SP}$. The theory of Ref. 17 reproduced well the features of the spectrum at $T>T_{SP}$.[4] The application of Raman scattering to probe the singlet excitations of two leg ladders is more recent.[7–9] 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,[18] 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

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

where $J>0$ and $J_\perp>0$ denotes the intrachain and interchain antiferromagnetic interactions, respectively. The interaction of light with the antiferromagnetic fluctuations is described by Loudon-Fleury's[12,13] photon-induced superexchange operator

$$H_R=\sum_{i,j} (\vec{E_I}\cdot\vec{\delta}_{ij})(\vec{E_S}\cdot\vec{\delta}_{ij})\vec{S_i}\cdot\vec{S_j}, \quad (2)$$

where $\vec{E}_I(\vec{E}_S)$ are the incident (scattered) electric field, and $\vec{\delta}_{ij}$ 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 Eq. (2) starting from the Hubbard Hamiltonian can be found in Ref. 19.

The Raman cross section[20,21] can be expressed as a function of the retarded Raman response function as

$$\frac{d^2\sigma}{d\Omega d\omega_2}=\frac{\omega_1\omega_2^3}{2\pi c^4 V}\frac{n_2}{n_1}\frac{1}{1-e^{-\beta\hbar\omega}}\mathrm{Im}\chi_R(\omega), \quad (3)$$

where ω_1 and ω_2 are the frequencies of the incoming and scattered radiation, respectively, and $\omega=\omega_2-\omega_1$, n_1, and n_2 are the respective refractive index. V is the volume of the crystal and c the velocity of light. The retarded linear response function $\chi_R(\omega)$ is defined as

$$\chi_R^{ret}(\omega)=\frac{i}{\hbar}\int_0^\infty e^{i(\omega+i0)t}\mathrm{Tr}\{Z^{-1}e^{-\beta H}[H_R(t),H_R(0)]\}, \quad (4)$$

where $Z=\mathrm{Tr}e^{-\beta H}$ and H_R is the Loudon-Fleury Hamiltonian (2).

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

$$\frac{d^2\sigma}{d\Omega d\omega_2} \propto \frac{1}{\hbar}\frac{1}{Z}\sum_{n,m} e^{-\beta E_n}|\langle\Psi_n|H_R|\Psi_m\rangle|^2 \times \delta[\omega-(E_n-E_m)/\hbar], \quad (5)$$

where $|\Psi_{n(m)}\rangle$ are eigenstates with energies $E_{n(m)}$. Such a formula can be easily interpreted as a Fermi golden rule averaged over the Boltzmann weight. To get information on two-magnons scattering processes we should perform a symmetry analysis of the matrix elements appearing in Eq. (5), 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 $2\pi/a$ (where a is the lattice spacing), spin, and its parity under leg exchange. The Raman operator defined in Eq. (2) is rotationally and translationally invariant, and still invariant under leg exchange. As a result, the selection rules impose that the states $|\Psi_n\rangle$ and $|\Psi_m\rangle$ 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 $\vec{E_I}$ and $\vec{E_S}$, parallel to the rung direction, thus we have

$$H_R = \frac{cste}{2} E_I E_S \sum_i \vec{S}_{1,i}\vec{S}_{2,i}. \quad (6)$$

In the following, we will evaluate the Raman intensity in the weak coupling and in the strong coupling limit using the standard Matsubara technique[22] to calculate the correlator $\chi_R(\omega)$.

To evaluate the time ordered Raman susceptibility for the weakly coupled chains, we will employ the Majorana fermion representation of the spin-ladder Hamiltonian (1) 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 $\xi^a_\nu(x)$ (ν=left,right) with bare mass $m_t=m=J_\perp$ and a singlet, $\rho_\nu(x)$ 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

$$H_\mu[\zeta] = \left[-i\frac{v_s}{2}\{\zeta_R\partial_x\zeta_R - \zeta_L\partial_x\zeta_L\} - i\mu\zeta_L\zeta_R\right], \quad (7)$$

where μ stands for the triplet or singlet mass and ζ 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

$$G^t_{\mu\nu}(k,i\omega_n) \equiv \langle \xi^\alpha_\mu(-\omega_n,k)\xi^\alpha_\nu(\omega_n,k)\rangle, \quad (8)$$

$$G^s_{\mu\nu}(k,i\omega_n) \equiv \langle \rho_\mu(-\omega_n,k)\rho_\nu(\omega_n,k)\rangle,$$

whose explicit expressions are

$$G^\alpha_{RR}(k,i\omega_n) = G^\alpha_{LL}(-k,i\omega_n) = -\frac{i\omega_n + v_s k}{\omega_n^2 + v_s^2k^2 + m_\alpha^2}, \quad (9)$$

$$G^\alpha_{RL}(k,i\omega_n) = G^\alpha_{LR}(k,i\omega_n)^* = -\frac{im_\alpha}{\omega_n^2 + v_s^2k^2 + m_\alpha^2},$$

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

$$H_R = \gamma_t\vec{\xi}_R\vec{\xi}_L + \gamma_s\rho_R\rho_L, \quad (10)$$

where $\gamma_t=m_t\gamma$ and $\gamma_s=m_s\gamma$. To arrive at this expression, the marginal term, already neglected in the derivation of the Hamiltonian (7), has been discarded. Injecting this expression into the definition of the Raman susceptibility (4) and applying Wick's theorem, the time ordered expectation value at finite temperature can be written as

$$\chi_R(i\omega_n) = \frac{1}{\beta}\sum_{\nu_n,\alpha}\gamma_\alpha^2\int\frac{dq}{2\pi}\{G^\alpha_{RL}(q,i\omega_n)G^\alpha_{LR}[-q,i(\omega_n - \nu_n)] - G^\alpha_{RR}(q,i\omega_n)G^\alpha_{LL}[-q,i(\omega_n-\nu_n)]\}. \quad (11)$$

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

$$\chi_R(i\omega_n) = \frac{1}{\beta}\sum_{\nu_n,\alpha}\gamma_\alpha^2\int\frac{dq}{2\pi} \times\left\{\frac{m_\alpha^2 - (i\nu_n + vq)[i(\omega_n-\nu_n)+vq]}{[\nu_n^2+(vq)^2+m_\alpha^2][(\omega_n-\nu_n)^2+(vq)^2+m_\alpha^2]}\right\}. \quad (12)$$

In order to evaluate the Matsubara sum in Eq. (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 $n_F(z)=1/[\exp(\beta z)+1]$ at the pole. Adding the four terms together yields

$$\chi_R(i\omega_n) = -\sum_\alpha \gamma_\alpha^2\int\frac{dq}{\varepsilon_\alpha(q)}\{1-2n_F[\varepsilon_\alpha(q)]\} \times 2vq\left[\frac{i\omega_n + 2vq}{\omega_n^2 + 4\varepsilon_\alpha(q)^2}\right], \quad (13)$$

where we have introduced the notation $\varepsilon_\alpha(q) = \sqrt{(vq)^2+m_\alpha^2}$. Thus performing the analytic continuation ($i\omega_n\to\omega+i0_+$), taking the imaginary part and performing the integral over q, we finally get

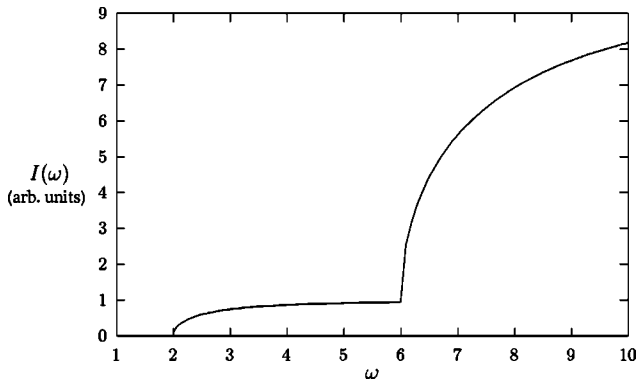

FIG. 1. Raman intensity in arbitrary units for $J_\perp \ll J$ at $T=0$ K obtained from the Majorana fermion approach. The frequency ω is measured in units of the gap.

$$\mathrm{Im}\chi_R(\omega)=\pi\sum_\alpha \tanh\left(\frac{\omega}{4k_BT}\right)\gamma_\alpha^2\frac{\sqrt{\omega^2-4m_\alpha^2}}{2\omega v}\Theta(|\omega|-2m_\alpha). \tag{14}$$

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 a result, the noninteracting Majorana fermions representation does not reproduce the Raman peak experimentally observed.[7–9] The spectra predicted by (14) is plotted on Fig. 1. The absence of signal for ω smaller than twice the gap is in qualitative agreement with numerical simulations.[18] 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 Gopalan, 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 $t_\alpha^\dagger$ ($\alpha=x,y,z$) 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

$$s^\dagger s+\sum_\alpha t_\alpha^\dagger t_\alpha=1 \tag{15}$$

has to be satisfied by the physical states. The representation of the spins $\mathbf{S}_1$ and $\mathbf{S}_2$ in terms of these singlet and triplet operators, is derived in Refs. 24,25. 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[25]

$$H_{\mathrm{MF}}=\left(\frac{J_\perp}{4}-\mu\right)\sum_{i,\alpha} t_{i,\alpha}^\dagger t_{i,\alpha}+\frac{Js^2}{2}\sum_{i,\alpha}(t_{i,\alpha}^\dagger+t_{i,\alpha})\times(t_{i+1,\alpha}^\dagger+t_{i+1,\alpha}). \tag{16}$$

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

$$G(k,i\omega_n)=-[\tilde{G}(k,i\omega_n)]^*=\frac{i\omega_n+\Lambda_k}{\omega_n^2+\omega_k^2}, \tag{17}$$

$$F(k,i\omega_n)=F^\dagger(k,i\omega_n)=\frac{2\Delta_k}{\omega_n^2+\omega_k^2},$$

where $\nu_n=2n\pi/\beta$ and the following notation has been introduced: $\omega_k^2=\Lambda_k^2-(2\Delta_k)^2$, with $\Delta_k=Js^2/2\cos k$ and $\Lambda_k=J_\perp/4-\mu+Js^2\cos k$, recovering the dispersion relation predicted by Gopalan, Rice, and Sigrist.[25] As shown in Ref. 25, the parameters μ 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 $\mathrm{Im}\chi_R(i\omega_n\rightarrow\omega+i0)$, where

$$\chi_R(i\omega_n)=\sum_{\alpha,\beta}\int_0^\beta d\tau e^{i\omega_n}\langle T_\tau(t_\alpha^\dagger t_\alpha)(\tau)(t_\beta^\dagger t_\beta)(0)\rangle. \tag{18}$$

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

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

Performing the usual linear response calculation,[22] we obtain as a final result:

$$\mathrm{Im}\chi_R(\omega)=\frac{\coth(\omega/4k_BT)[\omega/2(J_\perp/4-\mu)^2-1]^2}{4\omega\sqrt{[2Js^2/(J_\perp/4-\mu)]^2-[\omega/2(J_\perp/4-\mu)]^2-1]^2}}. \tag{20}$$

The Raman scattering spectra will show two peaks, one at energy $\omega=2\omega_\pi=2\Delta_s$ 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 ω^*, $I(\omega)\sim(\omega-\omega^*)^{-1/2}$. This behavior can be easily understood by a density of states argument. The resulting spectra is plotted in Fig. 2. No signal is obtained for $\omega<2\Delta_s$ in agreement with numerics.[18] Let us note that in recent experiments, a Raman scattering peak at twice the gap is observed in CaV_2O_5 (Ref. 8) where the spin-gap and the exchange constant are estimated to be $\Delta_s\sim 400\ \mathrm{cm}^{-1}$ and $J_\perp\sim 640$ K. These results are in qualitative agreement with our theory. In the case of $Sr_{14}Cu_{24}O_{41}$, the situation is more complicated due to the coexistence in the structure of dimerized spin chains, having a spin gap $\Delta_{\mathrm{chain}}=12$ meV (Ref. 26) and of spin ladders having a spin gap $\Delta_{\mathrm{ladder}}\simeq 32$ meV.[27] In Ref. 7,

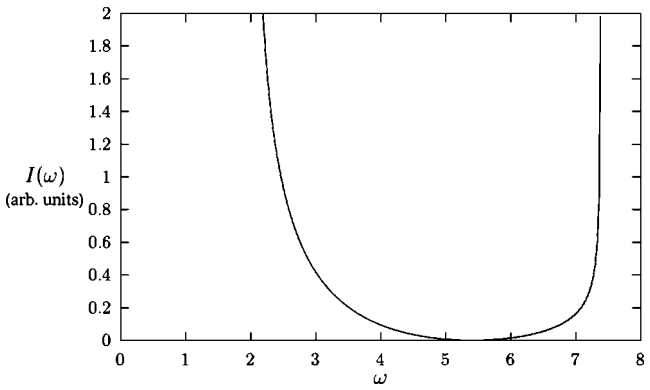

FIG. 2. Raman intensity in arbitrary units for $J_\perp/J=2$ at $T=0$ K. The frequency ω is measured in units of the gap.

a peak was obtained at 570 $cm^{-1} \simeq 71$ meV in Raman scattering experiments on polycrystalline samples. According to our theory, this would lead to a spin gap of $\simeq 35$ meV, in agreement with neutron scattering data. A more recent investigation or Raman scattering on single crystals[9] identifies a peak at 498 cm^{-1} as the Raman peak associated with the gap. The peak at 569 cm^{-1} is identified with a (0,0) gap. According to the authors of Ref. 9, 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.[28,29] In our treatment, we have been neglecting them altogether. They should give rise to peaks below the threshold 2Δ, as has been observed in experiments.[9] 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.

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