Transactions of the Materials Research Society of Japan 28 [4] 1307-1310 (2003)

Numerical Study of Magnetic Raman Spectra in Quantum Antiferromagnetic Systems

Yuhei NATSUME, Toshihiko SUZUKI and Nobuyuki KOIDE

Graduate School of Science and Technology, Chiba University, Inage-ku, Chiba 263-8522
Fax:81-43-290-2760, e-mail natsume@science.s.chiba-u.ac.jp

(Received)

According to the progress of synthesizing low-dimensional magnetic compounds, much attention has been paid to quantum antiferromagnets in which characteristic spin-gaps appear between singlet ground-states and triplet excited-states. In fact, magnetic Raman spectra in oxides compounds of CaV_2O_5 (so-called spin ladders) have been recently reported by Konstantinović *et al*, Phys. Rev. **B61**, 15185(2000). In the present work, we show the numerical reproduction of magnetic Raman spectra on the basis of exchange-scattering mechanism corresponding to these quantum antiferromagnetic materials. In calculation, the exact diagonalization and perturbation method are used. In CaV_2O_5, the observed characteristic features of line-shape at 795cm^{-1} has the quite asymmetric and broad structure. In this paper, we obtain the numerical results which agree quite well with those features without adjustable parameters. Furthermore, this method is applied to analysis of Raman spectra in $SrCu_2(BO_3)_2$, which is well confirmed as the Shastry-Sutherland model.
Key words: magnetic Raman spectra, exchange scattering, quantum antiferromagnets

§1. INTRODUCTION

Quantum antiferromagnets have no Néel ordering of magnetic moments, though the strong exchange interaction are contained between magnetic ions.

In fact, much attention has been paid to characteristic spin-gaps between singlet ground states and triplet excited states in these materials. As for these systems, the investigation of low-lying exited states including triplets should be made both on experimental and theoretical aspects [1,2] in order to understand essential properties caused by quantum magnetism. Here, we remark the spectra in oxides compounds of CaV_2O_5 (so-called spin ladders), which have been recently reported [3]. Corresponding such the experiments, we show the numerical reproduction of magnetic Raman spectra on the basis of exchange-scattering mechanism in this work. In calculation, the exact diagonalization and perturbation method are used. In CaV_2O_5, the observed characteristic features of line-shape which is quite asymmetric and broad have been reported [3]. Now, we get the numerical results which agree quite well with those features without adjustable parameters. On the basis of this work for CaV_2O_5, the present method of analyzing magnetic Raman spectra is adopted to those for new cupper compound $SrCu_2(BO_3)_2$, which is called Shastry-Sutherland Quantum Antiferromagetic model [4].

§2. THORY AND EXPERIMENT

In Raman spectra, the magnetic peaks are found in the quantum antiferromagnetic systems where the singlet ground states appear essentially accompanied by the singlet-triplet spin gappes. However, we would like to emphasize that the method of the explanation of those spectra for quantum antiferromagnetic systems with spin gap has not been confirmed up to now.

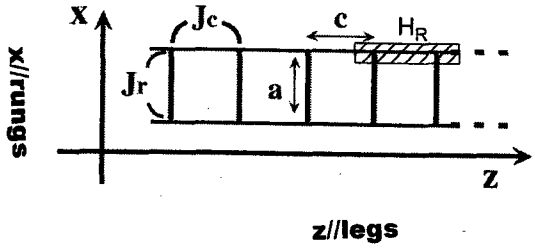
In fact, the experiment of Raman scattering in CaV_2O_5 which has antiferromagnetic ladder with two legs has been reported quite recently [3]. In this compound, the 1/2-spin is located on each lattice point of V^{4+}, i.e. this antiferromagnetic system is half-filled. In particular, the characteristic peaks due to magnetic scattering have been found in low temperature region of $T < 180K$ as shown in Fig.1.

The positions of such peaks are around 800cm^{-1}, which corresponds to twice the spin gap Δ_g = 562K= 398cm^{-1} [5,6]. Though the significant asymmetric shape for this peaks have been pointed out [3] in those spectra, the origin of the significant asymmetric shape has not been made clear up to now. In fact, the authors in [3] have tried to explain the shapes of peaks by the use of density of states (DOS) of magnon bands in one-dimensional systems. However, the shape of DOS is almost symmetric, because it closely reflects one-dimensional characters.

Therefore, we discuss, in this paper, the exchange-scattering spectra constructed by the perturbation method for J_c/J_r on the basis of the application of Loudon-Fleury theory [7] for magnetic Raman scattering. Such the application is shown schematically in Fig.2, where two adjacent singlet dimers turn into two adjacent triplet dimers by the scattering process of light [8,9].

§3. MODEL

Here, we consider a Heisenberg ladder with antiferromagnetic exchange coupling J_r for the rung and J_c for



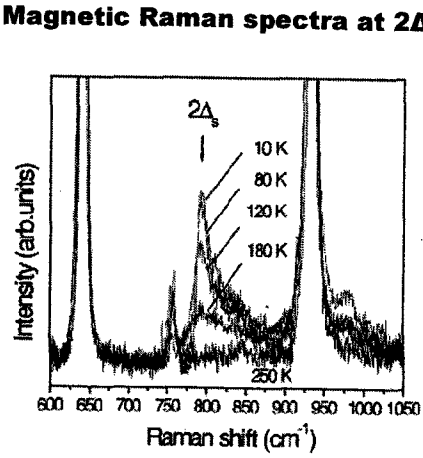


Fig. 1. Observed Raman spectra for CaV_2O_5 (ref.[2]) in the region of $600 \sim 1050cm^{-1}$. Quite an asymmetric shape of the peak asiggned as $2\Delta_g$ is found at $800 \sim 900\text{cm}^{-1}$. The schematic illustration of ladder system is also shown in the upper part.

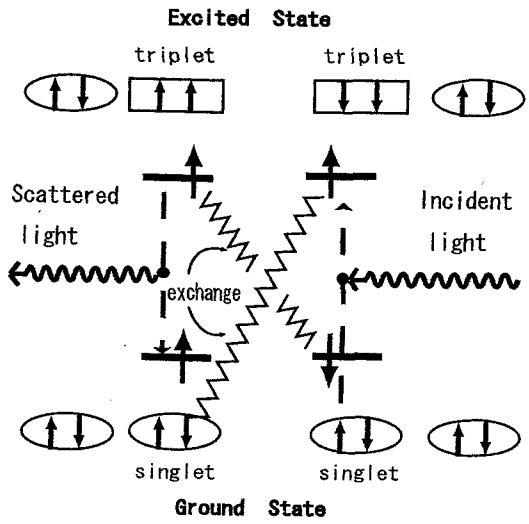


Fig. 2. The mechanism of the exchange scattering in the quantum antiferromagnetic systems. Two adjacent singlet dimers turn into two adjacent triplet dimers by the scattering process of light.

the leg. The present spin Hamiltonian is

$$\hat{H}_S^{\text{Ladder}} = J_r \sum_{<i,j>}^{\text{rung}} \hat{S}_i \cdot \hat{S}_j + J_c \sum_{<i',j'>}^{\text{leg}} \hat{S}_{i'} \cdot \hat{S}_{j'}, \quad (3.1)$$

where i and j denote sites at which spins $\hat{S}_i$ and $\hat{S}_j$ are located. Here, the summation is made for nearest neighbors on rungs and legs.

In this quantum antiferromagnetic system whose ground state is singlet, the so-called spin gap (Δ_g) corresponding to the transition from the singlet state to the triplet excited one appears. In cases of $J_c/J_r \sim 0.1$, the energy of this gap is estimated to be about around $0.9J_r$.

The Raman spectra are generally represented [10,11] as the dynamical Raman response function

$$I(\omega) = -\frac{1}{\pi} \cdot \text{Im}\langle\psi_0|\hat{H}_R^\dagger \cdot \frac{1}{D} \cdot \hat{H}_R|\psi_0\rangle, \quad (3.2)$$

where, $D = \omega + E_0 + i\delta - \hat{H}_S^{\text{Ladder}}$.
We consider that the temperature T is quite low in comparison with the spin gap Δ_g/k_B. Here, $\hat{H}_R$ called the radiation operator [7,10,11] expresses the interaction between lights and spins as

$$\hat{H}_R = \sum_{<i,j>} (\vec{E}_{inc} \cdot \vec{d}_{ij})(\vec{E}_{sc} \cdot \vec{d}_{ij})\hat{S}_i \cdot \hat{S}_j, \quad (3.3)$$

where $\vec{E}_{inc}$ and $\vec{E}_{sc}$ are polarization vectors of incident and scattered lights. Here, $\vec{d}_{ij}$ is the distance vector between i and j spin sites. The exchange interactions $\hat{S}_i \cdot \hat{S}_j$ play an important role to this scattering process. In $\hat{H}_R$, the summation means bonds for nearest neighbors of spin sites.

On the basis of results obtained by exact diagonalization in relatively small systems (spin-number=20), the perturbation method is adopted in large systems (spin-number=100) to the problem of the shape of peak for twice the spin gap. The detail of the procedure for the numerical calculation is discussed in other publications [8,9]. The exchange interaction J_c is adopted to the perturbation, because the ratio of J_c/J_r is nealy 0.1 in the present material. Energy positions and spectra are calculated by the second order perturbation.

§4. NUMERICAL RESULTS AND DISCUSSION

The calculated spectra for $(2\Delta_g)$ in the system of $N = 50 \times 2$ are shown in Fig.3. Here, the periodic boundary condition is adopted.

The agreement of spectra between the present calculation and the observed spectra at 795cm^{-1} in CaV_2O_5 reported by Konstantinović *et al.* [3] as shown in Fig.1 is quite well. As for this peak, we would like to point out that this peak should be treated as the transitions from the singlet ground state to the singlet excited states [14] apart from the traditional picture [7] of magnons [13,14].

In the ladder system for $J_c/J_r \ll 1$, the dimer ordering on rungs presents the almost adequate view of magnetic properties. In fact, we discuss the spectra obtained by the numerical calculation from the viewpoint of the transition from the singlet ground state which is

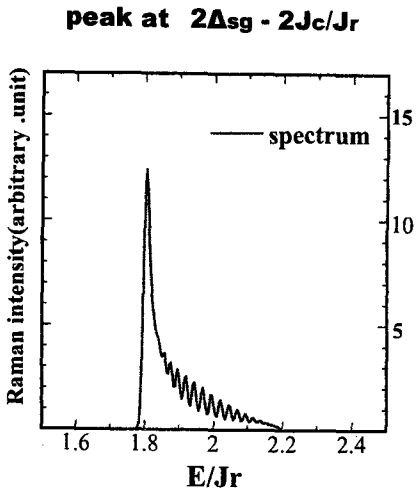


Fig. 3. The calculated shape for exchange-scattering spectra in the case of $J_c/J_r = 0.1$. The wavy shape is artificially caused by the width of δ in (3.2).

described as the dimer ordering on rungs. In addition to the condition of the optical transition between Γ points, the characters for properties in the symmetry of these excited levels corresponding to the Raman active peaks are assigned singlet composed of even number of triplets made by $\hat{H}_R$ in (3.3).

In order to discuss excited states corresponding to $2\Delta_g$, we adopt the bases where two triplet dimers are located on rungs whose number is ${}_{50}C_2 = 1225$. The transition by $\hat{H}_R$ of (3.3) from the ground state expressed as the ordering of singlet dimers on rungs occurs to bases in excited levels where the triplet dimers are made for adjacent rungs. In consideration of this property, we investigated patterns of triplet dimers on rungs in excited states. These patterns have been discussed in other publication [9]. In fact, comparing this result of the perturbation method and exact numerical diagonalization [1,2], we can show that the corresponding excited state mainly includes the triplet pairs on adjacent rungs. The intensity to this excited state is quite strong, because the Raman scattering is caused by $\hat{H}_R$ of (3.3) which has the exchange interaction for adjacent sits.

§5. SHASTRY-SUTHERLAND QUANTUM ANTIFERROMAGNETIC SYSTEMS

We proceed our calculation to the Exchange-Scattering Spectra in Shastry-Sutherland Quantum Antiferromagetic Systems [15,16]. The structure of antiferromagnetic bonds assigned as J_1 and J_2 is shown in the upper part of Fig.4.

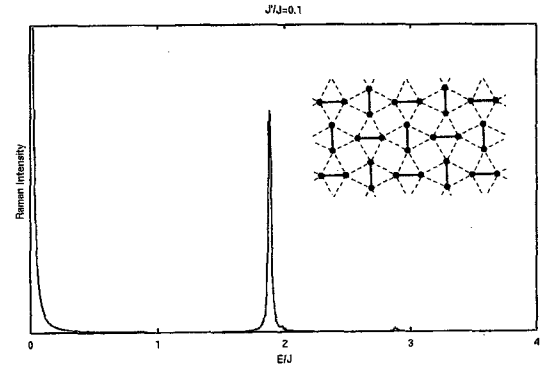


Fig. 4. Shastry-Sutherland model of quantum antiferromagetic systems is illustrated at the upper part, where solid and dashed lines mean antiferromagnetic bonds for J_1 and J_2. At the lower part, the calculated exchange-scattering spectra for J_2/J_1=0.1 is shown.

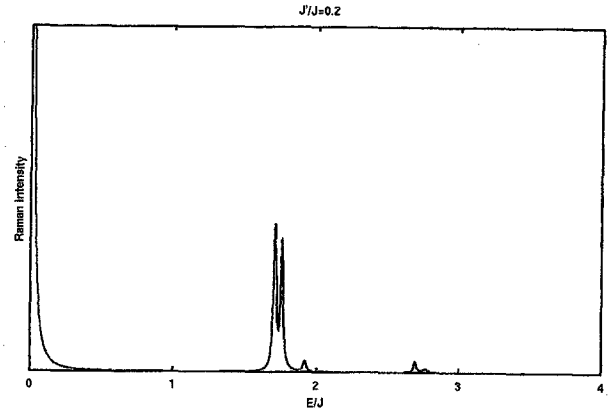


Fig. 5. The calculated spectra for J_2/J_1=0.2.

Here, the exchage interactions of the first nearest neighbors are antiferromagnetic expressed as J_1, while those for second nearest neighbors are also antiferromagnetic written as J_2. we discuss,in this paper, the calculated spectra by the use of the exact diagonalization method for various ratios of J_2/J_1 on the basis of theoretical investigation discussed in section 2.

In short, we consider a Heisenberg model, whose spin Hamiltonian is

$$\hat{H}_S^{S-S} = J_1 \sum_{<i,j>}^{1st\text{n.n.bonds}} \hat{S}_i \cdot \hat{S}_j + J_2 \sum_{<i',j'>}^{2nd\text{n.n.bonds}} \hat{S}_{i'} \cdot \hat{S}_{j'}, \tag{5.1}$$

where i and j denote sites at which spins $\hat{S}_i$ and $\hat{S}_j$ are located. In this quantum antiferromagnetic system whose ground state is singlet under the condition of $J_2/J_1 < 0.6$, the so-called spin gap (Δ_g) corresponding to the transition from the singlet state to the triplet excited one appears. [4,16,17,18,19] Needless to say, the properties of this system are dominated by the ratios of J_2/J_1.

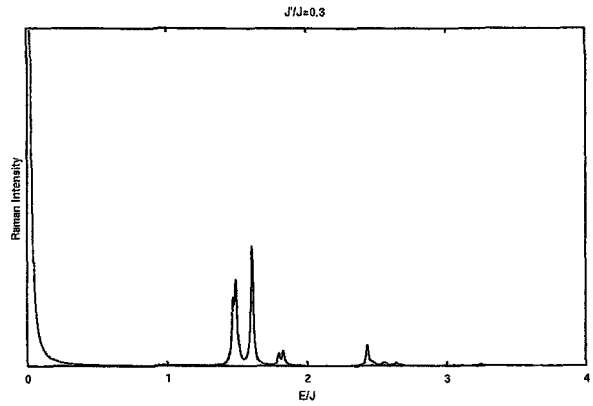


Fig. 6. The calculated spectra for $J_2/J_1 = 0.3$.

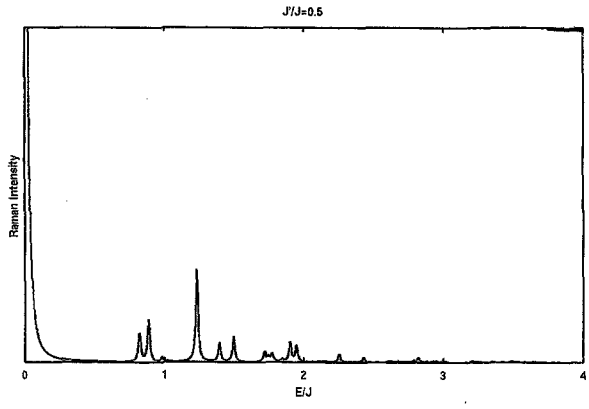


Fig. 7. The calculated spectra for $J_2/J_1 = 0.5$. In overall, we can find four structures corresponding to the observed ones in [4].

As a result of numerical study, we show the calculated spectra in Fig.4, 5,6 and 7, where values of J_2/J_1 are 0.1, 0.2, 0.3 and 0.5, respectively.

We suggest that the $2\Delta_g$ line clearly appears at $2J_1$ at $J_2/J_1 = 0.1$,while this line are spread into several ones around $J_1 < 2J_1$ at $J_2/J_1 = 0.5$. Furthermore, we would like to point out that the line of $3\Delta_g$ is get close to $2.2J_1$ with increasing J_2/J_1.

The comparison between present calculations and observed spectra [4] in $SrCu_2(BO_3)_2$ in relation with other several theoretical repots [16,17,18,19,20] for this model is the future subject.

Acknowledgements

We would like to thank Professor M. V. Mostovoy of the University of Groningen, Professor Y.Ueda and Dr. M.Isobe of Institute for Solid State Physics of the University of Tokyo, and Mr. S.Tada of Chiba University for the useful discussion. This work is supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Science, Sports and Culture. Numerical calculation in this work has been done using the facilities of the Supercomputer Center, Institute for Solid State Physics, University of Tokyo.

1) S. Tada, Y. Natsume and T.Suzuki, J. Phys. Soc. Jpn. **70**, 2443 (2001).
2) Y. Natsume, Y. Watabe and T. Suzuki, J. Phys. Soc. Jpn. **67**, 3314 (1998).
3) M.J.Konstantinović, Z.V.Popović, M.Isobe and Y.Ueda, Phys. Rev. **B61**, 15185 (2000).
4) P.Lemmens, M.Grove, M.Fisdher, G. Guntherodt, V. N. Kotov, H. Kageyama, K. Onizuka and Y.Ueda, Phys.Rev.Lett. **85**, 2605 (2000).
5) In the experimental work of neutron scattering, the spin gap is esitimated to be $500cm^{-1} \sim 700cm^{-1}$: R. S. Eccleston, M. Uehara, J. Akimitsu, H. Eisaki, N. Motokawa and S. Uchida, Phys. Rev. Lett., **81**, 1702 (1998).
6) On the basis of NMR-measurements of $1/T_1$ at the V sites, the estimation of $J_c = 93K, J_r = 655K, \Delta_g = 562K = 0.858J_r$ has been reported by the strong-coupling expansion within an independent magnon approximation: T. Ohama, M. Isobe and Y. Ueda, J. Phys. Soc. Jpn., **70**, 1801 (2001).
7) P.A. Fleury and R. Loudon, Phys. Rev. **166** (1968), 514.
8) Y. Natsume, S. Tada and T. Suzuki, J. Phys. Chem. Solids, **63**, 1361 (2002).
9) T. Suzuki, S. Tada and Y. Natsume, Prog. Theor. Phys. Suppl.**145**, 282 (2002).
10) K. Hibino and Y. Natsume, J. Phys. Soc. Jpn., **63**, 1630 (1994).
11) R.R.Singh, P.A.Fleury, K.B.Lyons, P.E.Sulewski, Phys. Rev. Lett., **62**, 2736 (1989).
12) Y. Watabe, S.Suzuki and Y.Natsume, Phys. Rev. **B52**, 3400 (1995).
13) Y. Watabe, T. Suzuki and Y. Natsume, J. Phys.: Condensed. Matter **6**, 7763 (1994).
14) M. Troyer, H. Tsunetsugu and D. Wurtz, Phys. Rev., **B 50**, 13515 (1994).
15) B.S.Shastry and B. Sutherland, Physica **108B**, 1069 (1981).
16) S.Miyahara and K.Ueda, Phys.Rev.Lett., **82**, 3701 (1999).
17) A.Koga and N.Kawakami, Phys.Rev.Lett., **84**, 4461 (2000),
18) C.Knetter, A. Buhler, E. Muller-Hartmann and G.S.Uhrig, Phys.Rev.Lett. **85**, 3958, (2000).
19) The perturbation method for the linkage of bonds is applied to this compound by Y.Fukumoto, J. Phys. Soc. Jpn., **69**, 2755, (2000).
20) The magnetization process is discussed for this compound by Y.Fukumoto, J. Phys. Soc. Jpn., **70**, 1397 (2001) :Y.Fukumoto and Akihide Oguchi, J. Mag. Mag. Mat, **226-230**, 612 (2001): Y.Fukumoto, J.Phys.Soc.Jpn., **69**, 1286 (2000).

Note added in proof— On the basis of the calculation for spin-correlation functions between sites for bonds J_1 and J_2, we have found the phase transition of the ground state at $0.6 < J_2/J_1 < 0.7$ in this Shastry-Sutherland quantum antiferromagnetic system.

(Received December 20, 2002; Accepted January 31, 2003)