Transactions of the Materials Research Society of Japan 29 [8] 3699-3702 (2004)

Study of Excited State in Quantum Antiferromagnets by Computational Reproduction of Magnetic Raman Spectra

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According to the progress of synthesizing low-dimensional magnetic compounds, much attention has been paid to quantum antiferromagnets (spin=1/2) in which characteristic spin-gaps appear between singlet ground-states and triplet excited-states. In fact, magnetic Raman spectra in oxide compounds of $SrCu_2(BO_3)_2$ have been recently reported by Lemmens *et al.* Phys. Rev. Lett. **85**, 2605 (2000). In the present work, we show the numerical reproduction of magnetic Raman spectra on the basis of exchange-scattering mechanism making correspondence to these orthogonal-dimer-type quantum antiferromagnetic systems. In calculation, the exact diagonalization are used. In $SrCu_2(BO_3)_2$, the observed characteristic features of appearing dominant four lines have been reported. In this paper, we obtain the numerical results which agree qualitatively with those features without adjustable parameters. In consideration that this $SrCu_2(BO_3)_2$ is well confirmed as the Shastry-Sutherland model proposed theoretically, we investigate correlation functions between spins for cases with various ratios of exchange interactions. In fact, we discuss phases of dimers, plaquettes and square lattice antiferromagnets.
Key words: magnetic Raman spectra, exchange scattering, quantum antiferromagnets

1. INTRODUCTION

Quantum antiferromagnets have no Neel ordering of magnetic moments, though the strong exchange interaction is 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 with S=1/2. As for these systems, the investigation of low-lying excited states including triplets should be made both on experimental and theoretical aspects [1,2] in order to understand essential properties caused by quantum magnetism. In several years, we have discussed the numerical reproduction of magnetic Raman spectra on the basis of exchange-scattering mechanism for those quantum anitferromagnets [3-7]. In the present paper, we show the analysis of magnetic Raman spectra for new copper compound $SrCu_2(BO_3)_2$ [8], in which orthogonal dimers are distributed on the plane. This structure is schematically illustrated in Fig.1.

It should be noted that this is also called Shastry-Sutherland quantum antiferromagetic model [9]. In addition to the discussion of exchange scattering spectra, the behavior of correlations between spins is studied.

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 gaps. 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.

Therefore, we discuss, in this paper, the numerical reproduction of the exchange-scattering spectra on the basis of the application of Loudon-Fleury theory [10] for magnetic Raman scattering.

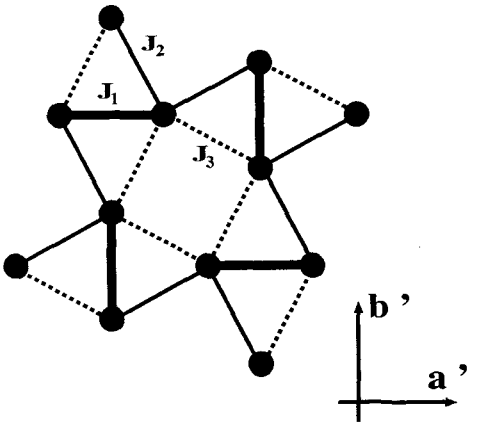

Fig. 1. The spin structure of $SrCu_2(BO_3)_2$ [8]. The crystal axes of a' and b' on the c-plane are indicated. The orthogonal dimers are distributed on the plane.

2. MODEL

Here, we consider a Heisenberg antiferromagnets with antiferromagnetic exchange coupling J_{ij} for each bond. The present spin Hamiltonian is

$$\hat{H}_S = \sum_{<i,j>} J_{ij} \hat{S}_i \cdot \hat{S}_j \qquad (1)$$

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, the so-called spin gap (Δ_g) corresponding to the transition from the singlet state to the triplet excited one appears.

The Raman spectra are generally represented [10,11,12] as the dynamical Raman response function

$$I(\omega) = -\frac{1}{\pi} \times \mathrm{Im}\langle\psi_0|\hat{H}_R^{\dagger}\frac{1}{\omega + E_0 + i\delta - \hat{H}_S}\hat{H}_R|\psi_0\rangle, \quad (2)$$

where ψ_0 is the ground state. 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 is the interaction between lights and spins as

$$\hat{H}_R = \sum(\vec{E}_{inc}\cdot\vec{d}_{ij})(\vec{E}_{sc}\cdot\vec{d}_{ij})\hat{S}_i\cdot\hat{S}_j, \quad (3)$$

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

3. SHASTRY-SUTHERLAND QUANTUM ANTIFERROMAGNETIC SYSTEMS

We discuss the calculation of the exchange-scattering spectra in Shastry-Sutherland quantum antiferromagetic systems. The structure of antiferromagnetic bonds assigned as J_1, J_2 and J_3 is shown in Fig.1.

Here, the exchange interactions of the first nearest neighbors are antiferromagnetic and expressed as J_1, while those for second nearest neighbors are also antiferromagnetic and written as J_2 and J_3. We discuss the calculated spectra by the use of the exact diagonalization method for various ratios of J_2/J_1 in cases of $J_2 = J_3$ on the basis of theoretical investigation discussed in previous section.

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

$$\hat{H}_S^{S-S} = J_1\sum^{1st}\hat{S}_i\cdot\hat{S}_j + J_2\sum^{2nd}\hat{S}_{i'}\cdot\hat{S}_{j'} + J_3\sum^{2nd}\hat{S}_{i'}\cdot\hat{S}_{j'}, \quad (4)$$

In this quantum antiferromagnetic system whose ground state is singlet under the condition of $J_2/J_1 < 0.667$ [13], the so-called spin gap (Δ_g) corresponding to the transition from the singlet state to the triplet excited one appears. [13,14,15] Needless to say, the properties of this system are dominated by the ratios of J_2/J_1.

4. EXCHANGE-SCATTERINF SPECTRA

As a result of numerical study, we show the calculated spectra in Fig.2, where values of J_2/J_1 are 0.1, 0.2, 0.3, 0.4, 0.51 and 0.6, 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 $J_2/J_1 = 0.4$. Furthermore, we would like to point out that the line of $3\Delta_g$ gets close to $2.2J_1$ with increasing J_2/J_1. In the case of $J_2/J_1 = 0.6$, we

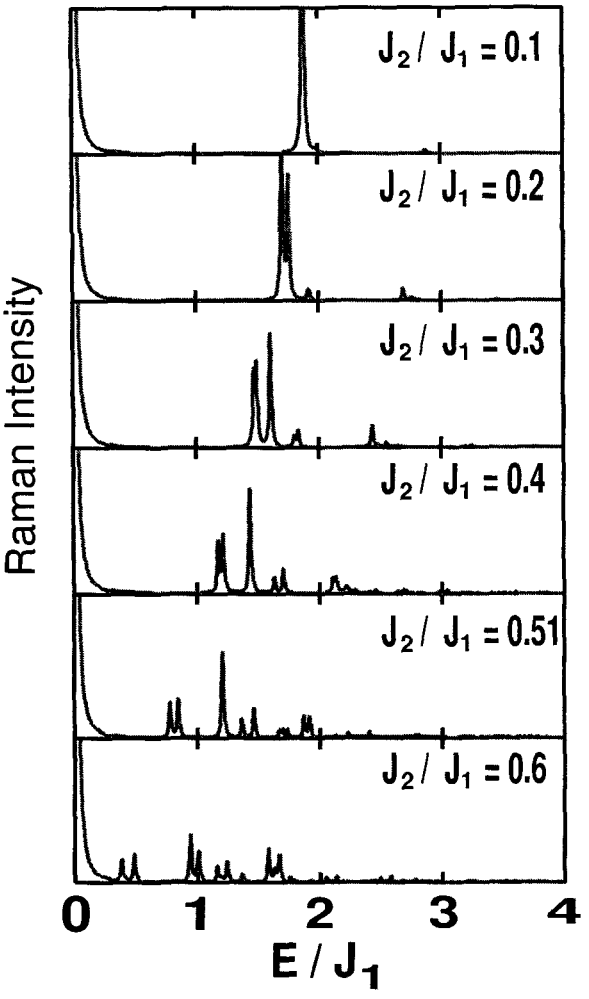

Fig. 2. Calculated exchange scattering spectra. The values of J_2/J_1 are 0.1, 0.2, 0.3, 0.4, 0.51 and 0.6, respectively.

can find four lines in spectra. This feature qualitatively agrees with that of experiments [8]. The quantitative discussion is the future subject [16,17].

5. CORRELATIONS BETWEEN SPINS

In order to investigate the relation between various magnetic phases in the present model, such as dimer states, plaquette antiferromagnets and square lattice antiferromagnets, we show calculated results of correlation functions between spins in this section. The correlation function is defined as $\langle S_{iz}\cdot S_{jz}\rangle_{J_\ell}$, where S_i and S_j are located at edges of J_ℓ bonds. Namely, ferromagnetic and antiferromagnetic correlations are reflected by positive and negative values of correlation functions, respectively. In the left column of Fig.3, correlations for J_1 and J_2 are illustrated in the case of $J_3 = J_2$. Here, the axis of abscissa is J_3/J_1, i.e. J_2/J_1. The antiferromagnetic correlations for J_1 bond appearing in $J_3/J_1 < 0.6$ suggest the dimer state on J_1 bond. On the other hand, the ground state turns to square lattice antiferromagnetic one in $J_3/J_1 \geq 0.7$. Accordingly, the ferromagnetic correlation is found on J_1 bond. In correspondence with

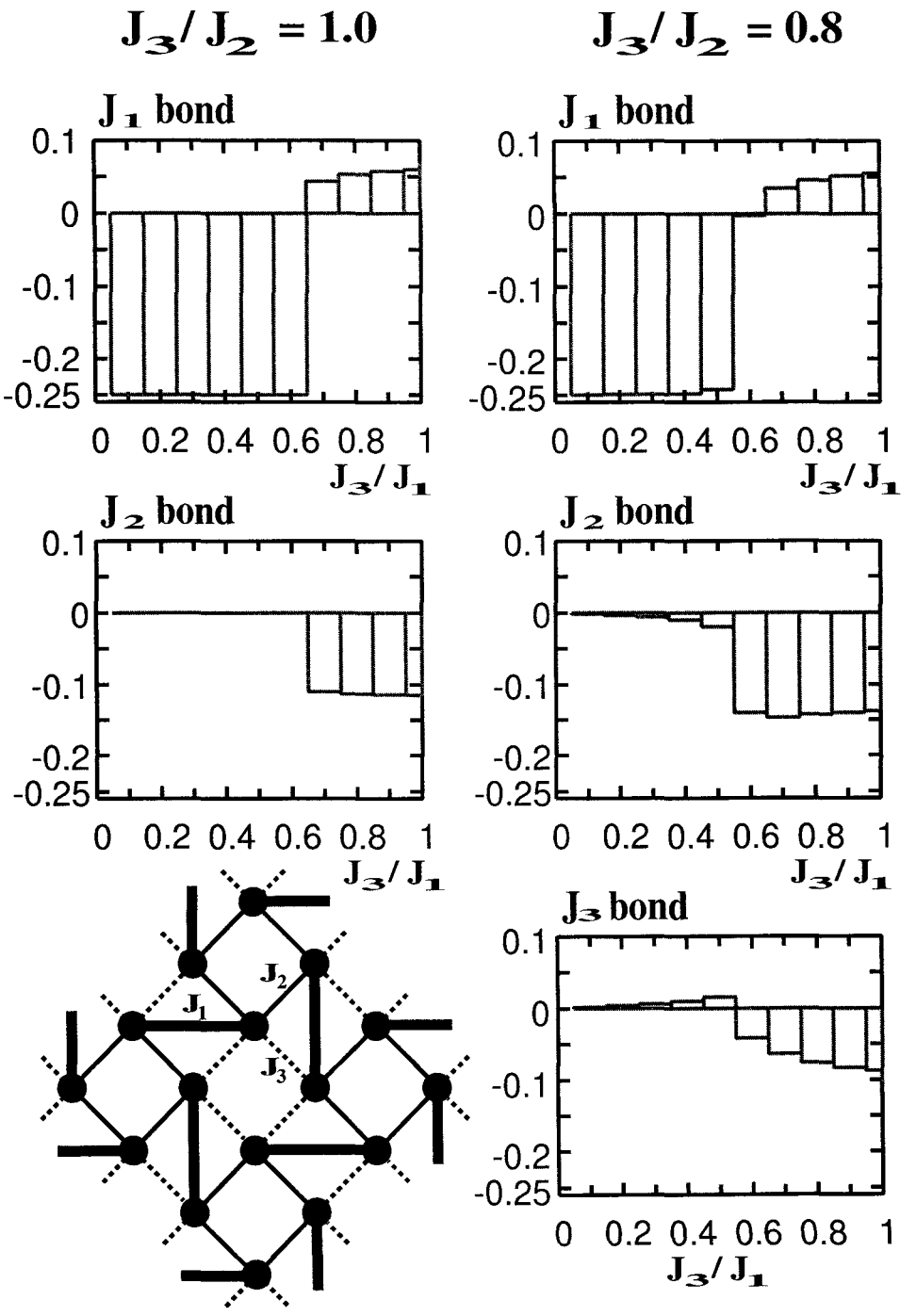

Fig. 3. Calculated correlation functions between spins. In the left column, those for J_1 and J_2 are illustrated in the case of $J_3 = J_2$. In the right column, those in the case of $J_3/J_2 = 0.8$ are shown.

this behavior, antiferromagnetic correlations grow up on J_2 and J_3 bonds.

In the next place, we concentrate our attention on correlations in the case of $J_3/J_2 = 0.8$, which are shown in the right column of Fig.3. It should be noted that the behavior of correlations for J_2 bond is quite different from that of J_3 bond. As for J_1 bond, the strength of antiferromagnetic correlation is suppressed around $J_3/J_1 \approx 0.6$, though the transition from the dimer state to the square lattice antiferromagnet appears. This transient features reflect the plaquette state, where the antiferromagnetic plaqettes composed of J_2 bonds are formed. In fact, the correlation for J_2 bond is enhanced about $J_3/J_1 = 0.6$. Further, we can find the peak of antiferromagnetic correlation of J_2 bond at $J_3/J_1 = 0.7$. In accordance with this property for J_2 bond, we can see the remarkable behavior for J_3 bond in the lower part of Fig.3: the correlation changes from ferromagnetic characters to antiferromagnetic ones. The weakness of the correlation for J_3 bond around $0.5 < J_3/J_1 < 0.7$ suggests the plaquette states composed of J_2 bonds.

Acknowledgements

We would like to thank Professor M. V. Mostovoy of the University of Groningen, Dr. Y. Fukumoto of Tokyo University of Science, 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.

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(Received October 13, 2003; Accepted April 24, 2004)