Theory of $L$-edge resonant inelastic x-ray scattering for magnetic excitations in two-leg spin ladder

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(Dated: May 1, 2014)

We study the magnetic excitation spectra of Cu $L_{2,3}$-edge resonant inelastic x-ray scattering (RIXS) from the spin-liquid ground state in two-leg spin ladder cuprates. Applying the projection method developed by the present authors, we have derived the formulas of the magnetic RIXS spectra, which are expressed by one- and two-spin correlation functions in the polarization changing and preserving channels, respectively. The one-spin correlation function includes both one- and two-triplon excitations and they are picked up separately by choosing rung wave vector $-q_a = \pi$ and 0, respectively. An application to Sr$_{14}$Cu$_{23}$O$_{41}$ reveals that the calculated RIXS spectrum captures well the dispersive behavior shown by the lower boundary of two-triplon continuum. By adjusting the geometrical configuration of the measurement, one-triplon dispersion around the zone center could be detectable in the RIXS experiment. The observed weak intensity in the higher energy region might be attributed to the two-spin correlation function, which could be detected more clearly at the $M_3$-edge RIXS spectra.

PACS numbers: 78.70.Ck, 72.10.Di, 78.20.Bh, 74.72.Cj

I. INTRODUCTION

Resonant inelastic x-ray scattering (RIXS) provides us with one of the rare opportunities to investigate various excitations in solids including charge, orbital, and spin degrees of freedoms. Among them, extracting the magnetic excitation by RIXS is rather challenging because to distinguish its intensity from other contributions such as the elastic signal and phonon excitations is hard to achieve due to the small energy transfer below 1 eV associated with the magnetic excitation. However, it is worthwhile to pursue potential of RIXS as a promising probe for magnetic excitation, since it can survey a wide range of momentum transfer in the Brillouin zone and requires only small sample volume. These features convince us RIXS to be a choice complementary to the conventional inelastic neutron scattering (INS).

Recently, progress in the experimental technique of RIXS enables us to carry out higher energy resolution measurement for instance, in several transition metal oxides like the cuprates. Hill et al. have succeeded in detecting the magnetic excitation spectra peaked around 500 meV energy region at the Cu $K$-edge in La$_2$CuO$_4$. The following theories have revealed that the obtained signals are attributed to the two-magnon excitation from the antiferromagnetic (AFM) ground state brought about by the presence of the core hole potential during the intermediate states. Since then, plenty of measurements have been reported out at the Cu $K$-edge as well as Cu $L_{3}$-edge.

When we turn our attention to the $L$-edge RIXS, the situation becomes a little complicated. In the Cu $L$-edge RIXS process, the transition is between the $2p$ core and $3d$ states. The photo-excited electron eliminates the 3$d$ hole leading the 3$d$ state to the closed shell. The absence of the spin degree of freedom at the core-hole site is similar to the situation of the non-magnetic impurity problem in the spin system. Constructing a relevant theory to handle such difficult situation is challenging but attracting. Within the fast collision approximation, the momentum and polarization dependences of the magnetic excitation spectra at the Cu $L_{2,3}$-edges were investigated. In another attempts, resonant energy dependence of the RIXS spectra is described by the scattering operator inferred from elastic scattering.

In order to extract the magnetic excitation from the closed 3$d$ state at the core hole site in the intermediate state, we have developed an effective theory to investigate the magnetic RIXS spectrum at the Cu $L_{2,3}$ edges. The theory treats the polarization and energy dependences of the spectrum faithfully, projecting the final states onto the possible spin excited states, from which the RIXS spectra are expressed as the form of the spin correlation functions. They consist of the two-spin as well as one-spin correlation functions. The latter is also found in the theory of INS, but the former is specific to RIXS. Even for the one-spin correlation function, its transferred momentum dependence turns out to be completely different from the one in the INS theory reflecting the inclusion of the non-local magnetic excitation.

We have applied our theory to two-dimensional system having the AFM ground state and confirmed the quantitative effectiveness of the theory. It has reproduced well the experimental data reported by Guarise et al. in Sr$_2$CuO$_2$Cl$_2$. Contributions of the multi-magnon excitations have been evaluated too. Then, we have applied our theory to one-dimensional Heisenberg chain having the spin-singlet ground state that preserves rotational.
invariance in the spin space. The RIXS spectrum, which can cover the magnetic excitations generated not only at the core-hole site but also around the core-hole site, has been derived in a way that manifests the rotational invariance. The results have been consistent with those obtained by other theoretical approaches. Our result also suggests that contribution from the two-spin correlation function is detectable at the higher energy region in the \( \sigma \)-polarization measurement. Unfortunately, at that time, no experimental data was available to compare with our results.

In this work, we extend our theory to exploit the Cu \( L_{2,3} \)-edge magnetic excitation spectrum in RIXS for two-leg spin ladder cuprate where the ground state is the spin-liquid retaining the spin rotational invariance. The low energy sector of the spin excitations are known as one- and two-triplon excitations. Our analysis reveals that the RIXS spectra can trace the one- and two-triplon dispersions as a function of the transferred momentum through the one-spin correlation function.

Then, we apply our theory to interpret the experimental results reported by Schlappa et al. in \( \text{Sr}_{14-\tau}\text{Ca}_\tau\text{Cu}_{24}\text{O}_{41} \) \( \tau \) undoped \( \text{Cu} \) Ladder system \( \text{Sr}_{14}\text{Cu}_{24}\text{O}_{41} \). This material is considered an experimental realization of a two-leg ladder structure (Fig. (a)) and has attracted of particular interest since Cu doped systems \( \text{Sr}_{14-\tau}\text{Ca}_\tau\text{Cu}_{24}\text{O}_{41} \) have exhibited superconducting state under pressure for \( x \) ranging from 11.5 to 13.6. In the RIXS measurement at the Cu \( L_3 \)-edge (930.6 eV), Schlappa et al. observed the dispersive signals attributed to the lower boundary of two-triplon excitation continuum. Our theory succeeds in reproducing the momentum dependence of the RIXS spectral profile. In addition, we find one-triplon dispersion can be detected by rotating the sample around the \( b \)-axis. The observed weak intensity in the higher energy region might be the contribution from the two-spin correlation function, mainly originated from two-triplon excitations with total spin \( S = 0 \). We argue that the contribution from the pure two-spin correlation function is distinguishable more clearly at the \( M_3 \)-edge measurement.

The present paper is organized as follows. In Sec. II we extend the theory developed in our previous papers aiming at the application to the Cu \( L_{2,3} \) edges in two-leg ladder systems. The RIXS spectra are expressed in terms of spin-correlation functions. In Sec. III the amplitudes leading to the spin-correlation functions are evaluated on a finite-size two-leg ladder cluster. An application to \( \text{Sr}_{14}\text{Cu}_{24}\text{O}_{41} \) is shown in Sec. IV. Section V is devoted to summary and discussion.

II. THEORETICAL FRAMEWORK

A. Initial state and magnetic excitations

For the purpose of application to the two-leg spin ladder system \( \text{Sr}_{14}\text{Cu}_{24}\text{O}_{41} \), we consider the system is at half-filling and is undoped with each Cu atom having one hole per site in the \( x^2 - y^2 \) orbital. The \( x \) and \( y \) axes are defined along the Cu-O bonds parallel to the crystallographic \( c \) and \( a \) axes, respectively, while the \( z \) along the \( b \) axis. Here, the \( a \) and \( c \) axes are along the rung and leg directions of the ladders. The ground state and the low-energy spin excitations are described by the \( S = 1/2 \) antiferromagnetic Heisenberg Hamiltonian with an additional four-spin exchange terms.

\[
H_{\text{mag}} = J_{\parallel} \sum_{i} \sum_{\tau = 1}^{2} \mathbf{S}_{i,\tau} \cdot \mathbf{S}_{i+1,\tau} + J_{\perp} \sum_{i} \mathbf{S}_{i,\tau} \cdot \mathbf{S}_{i,\tau+1} + J_{\text{cyc}} \sum_{\text{plaquette}} \left( \mathbf{S}_{i,\tau} \cdot \mathbf{S}_{i+1,\tau} \right) \left( \mathbf{S}_{i+1,\tau} \cdot \mathbf{S}_{i+1,\tau+1} \right) - J_{\text{cyc}} \sum_{\text{plaquette}} \left( \mathbf{S}_{i,\tau} \cdot \mathbf{S}_{i+1,\tau} \right) \left( \mathbf{S}_{i+1,\tau} \cdot \mathbf{S}_{i,\tau+1} \right),
\]

where the index \( i \) refers to the rungs. The symbol \( \tau \in \{1, 2\} \) discriminates legs, and \( \tau \) denotes 2 for \( \tau = 1 \), and vice versa. The exchange coupling constants along the legs and rungs are denoted as \( J_{\parallel} \) and \( J_{\perp} \), respectively (Fig. (b)). In addition, four-spin coupling \( J_{\text{cyc}} \) is included.
We briefly summarize the ground state and low energy sector of the spin excitations of \( H_{\text{mag}} \). The ground state is known to be the gapped spin liquid due to the quantum fluctuation.\(^{25,32}\) Unlike the AFM ordered state, the spin-liquid state preserves the rotational invariance. First, let us assume \( J_{\parallel} = J_{\text{cyc}} = 0 \). Since each rung is independent, the ground state is constructed by \( N/2 \) pairs of rung singlet where \( N \) denotes the number of spins. This state is called as rung singlet.\(^{26,37}\) The lowest energy excited state is realized by changing one of the singlets into triplet with the excitation energy, or equivalently a spin gap, \( J_{\perp} \). As the number of triplets increases, the excited states are called as one-, two-, three-triplon excitations, and so on.

When \( J_{\parallel} \) is turned on, this kind of simple picture may be invalid. However, it is known that even when \( J_{\parallel} \rightarrow \infty \), the ground state is adiabatically connected to the rung singlet.\(^{40}\) Thus, classifying the magnetic excitation by the number of triplons gives a good description and the spin gap remains finite.\(^{28,35–39}\) The presence of \( J_{\text{cyc}} \) does not alter the nature of the ground and low energy excited states qualitatively for \( J_{\text{cyc}}/J_{\perp} \sim 0.25 \), although complication sets in when the ratio grows, which is beyond the present interest.\(^{41}\)

### B. RIXS spectra

We extend our RIXS theory developed for spin-singlet one-dimensional Heisenberg chain to quasi one-dimensional two-leg spin ladder system showing the spin-liquid ground state. In the following, a concise version of the explanation of the theory is displayed, relegating a detail to ref.\(^{24}\)

In the electric dipole (E1) transition at the transition-metal \( L_{2,3} \)-edge, the transition progresses between the 2p-core state and the 3d state. This process, described by the electron-photon interaction Hamiltonian \( H_{\text{int}} \), is mediated by absorbing the incident photon with wave vector \( \mathbf{q} \) and energy \( \omega_i \), and then, emitting the scattered photon with wave vector \( \mathbf{q} \) and energy \( \omega_f \). The RIXS spectra may be expressed by the second-order E1 allowed process.\(^{23,24}\)

\[
W(q_f \alpha_f; q_i \alpha_i) = 2\pi \sum_{f'} \sum_n \frac{\langle \Phi_f' | H_{\text{int}} | n \rangle \langle n | H_{\text{int}} | \Phi_i \rangle^2}{E_g + \omega_i - E_n + i\Gamma} \times \delta(E_g + \omega_i - E_{f'} - \omega_f),
\]

with \( q_i \equiv (\mathbf{q}, \omega_i), q_f \equiv (\mathbf{q}_f, \omega_f), |\Phi_i\rangle = c_{\mathbf{q}_i \alpha_i} |g\rangle, |\Phi_f\rangle = c_{\mathbf{q}_f \alpha_f} |f\rangle \), where \( |g\rangle \) and \( |f\rangle \) represent the ground state and excited states of the matter with energy \( E_g \) and \( E_f \), respectively. The polarization directions of the incident and scattered photons are \( \alpha_i \) and \( \alpha_f \), respectively. The annihilation (creation) operator of photon with momentum \( \mathbf{q} \) and polarization \( \alpha \) is denoted as \( c_{\mathbf{q} \alpha} \). The intermediate state is denoted as \( |n\rangle \) with energy \( E_n \) in the presence of the core-hole. Since \( E_n \) includes the core-hole energy \( \epsilon_{\text{core}} \), we express \( E_n = \epsilon_{\text{core}} - i\Gamma + \epsilon_n \) where \( \Gamma \) stands for the lifetime broadening width of the core-hole and \( \epsilon_n \) is the energy of the spin part in the intermediate state. The \( \epsilon_n \) will be evaluated by the Hamiltonian \( H_{\text{mag}} \); constructed from \( H_{\text{mag}} \) by eliminating the spin degree of freedom at the central core-hole site.

In the intermediate state, the spin degree of freedom is lost at the core-hole site. The final state experienced such intermediate state may be expressed by \( |S_{0,1}(g), S_{0,2}(g), S_{1,1}(g), \cdots \rangle \) in the polarization changing channel, and \( |S_{0,1}, S_{0,2}(g), \cdots \rangle \) in the polarization preserving channel, where suffix (0, 1) indicates the core-hole site. Paying attention to the non-orthogonality of these state, we project the final state on these state in the same way as carried out in our previous study on the one-dimensional system. We obtain

\[
\sum_n \frac{H_{\text{int}} |n\rangle \langle n| H_{\text{int}} |\Phi_i\rangle}{E_g + \omega_i - \epsilon_{\text{core}} - \epsilon_n + i\Gamma} = \left(-\frac{i}{15}\right) \alpha_f \times \alpha_i \cdot \left[ f_1^{(1)}(\omega_i) S_{0,1} \right.
+
\left. f_2^{(1)}(\omega_i) (S_{1,1} + S_{-1,1}) + f_3^{(1)}(\omega_i) S_{0,2} \right.
+
\left. f_4^{(1)}(\omega_i) (S_{1,2} + S_{-1,2}) \right]\]|g\rangle,
\]

(2.3)

where \( \alpha_{f\perp} \) and \( \alpha_{f\parallel} \) stand for the polarization vectors of the incident and scattered photons, respectively, projected onto the \( xy \) plane. The \( f_p^{(n)}(\omega_i) \)'s are the coefficients to be determined. They could be accurately evaluated in a system having rather small size, since the relevant excited states are restricted around the core-hole site.

Accordingly, the RIXS spectra for the polarizations \( \alpha_i(f) = (\alpha_i^x(f), \alpha_i^y(f), \alpha_i^z(f)) \) are expressed as

\[
W(q_f \alpha_f; q_i \alpha_i) \propto \left( \frac{\alpha_f^x \alpha_i^y - \alpha_f^y \alpha_i^x}{15} \right)^2 Y^{(1)}(\omega_i; q_c, q_a, \omega)
+
\left( \frac{2 \left( \alpha_f^x \alpha_i^x + \alpha_f^y \alpha_i^y \right)}{15} \right)^2 Y^{(2)}(\omega_i; q_c, q_a, \omega),
\]

(2.4)

where the first and second terms represent the contributions from the polarization changing and preserving channels, respectively. The \( Y^{(1)}(\omega_i; q_c, q_a, \omega) \) and \( Y^{(2)}(\omega_i; q_c, q_a, \omega) \) are Fourier transforms of the one-spin
and two-spin correlation functions defined by
\[ Y^{(1)}(\omega; q_c, q_a, \omega) = \int \langle Z^{(1)}(\omega; q_c, q_a, t) Z^{(1)}(\omega; q_c, q_a, 0) \rangle e^{i\omega t} dt \]
\[ Y^{(2)}(\omega; q_c, q_a, \omega) = \int \langle Z^{(2)}(\omega; q_c, q_a, t) Z^{(2)}(\omega; q_c, q_a, 0) \rangle e^{i\omega t} dt \]
with
\[ Z^{(1)}(\omega; q_c, q_a) = \sum_{j,\tau} e^{-iV_{j,\tau} - iV_{q_c, q_a}(\tau - 1)a_r}
\times [f_1^{(1)}(\omega)S_{\tau,\tau} + f_2^{(1)}(\omega)(S_{\tau+1,\tau} + S_{\tau+1,\tau}^{-1})
+ f_3^{(1)}(\omega)(S_{\tau,\tau} + f_4^{(1)}(\omega)(S_{\tau+1,\tau} + S_{\tau+1,\tau}^{-1})], \quad (2.7)
\]
\[ Z^{(2)}(\omega; q_c, q_a) = \sum_{j,\tau} e^{-iV_{j,\tau} + iV_{q_c, q_a}(\tau - 1)a_r}
\times [f_2^{(2)}(\omega)(S_{\tau+1,\tau} + S_{\tau-1,\tau}) + f_3^{(2)}(\omega)S_{\tau,\tau}
+ f_4^{(2)}(\omega)(S_{\tau+1,\tau} + S_{\tau-1,\tau})] \cdot S_{\tau,\tau}. \quad (2.8) \]

The coordinate of the site at the \(j\)-th rung and \(\tau\)-th leg is denoted as \(r_{j,\tau} = (\tau - 1)a_r\) in the \(ca\) plane. The symbols \(q_c\) and \(q_a\) represent the wave numbers along the leg and rung directions, respectively. They are the corresponding components of vector \(q\), which is the projection of the scattering vector \(q = q_i - q_f\) onto the \(ca\)-plane. In the two-leg ladder configuration, the latter takes two relevant values 0 and \(\pi/a_r\). Hereafter, the momenta \(q_c\) and \(q_a\) are measured in units of 1/\(c_L\) and 1/\(a_r\), respectively, when their numerical values are mentioned.

Note that in the above expressions (2.7) and (2.8), the magnetic excitations included are those at the neighboring sites within the two adjacent plaquettes linked to the central site as well as at the central core-hole site. Notice also that if the fast collision approximation is adopted, \(Z^{(1)}(\omega; q_c, q_a)\) and \(Z^{(2)}(\omega; q_c, q_a)\) become \(f_1^{(1)}(\omega)\sum_{s,\tau} e^{-iV_{s,\tau} - iV_{q_c, q_a}(\tau - 1)a_r} S_{\tau,\tau}^z\) and 0, respectively, meaning that only the excitation at the core-hole site is relevant to the former while the latter vanishes.

The first term of Eq. (2.4) gives the spectral shape as a function of \(\omega\) similar to the conventional one-spin correlation function familiar to INS theory: the presence of \(f_2^{(1)}(\omega)\) and \(f_3^{(1)}(\omega)\) modifies the \(q_c\) dependence of the spectral intensity. The complicated \(q_a\) dependence is a direct consequence of the inclusion of non-local spin excitation around the core-hole site as seen from Eq. (2.7), which is missing in the fast collision approximation.

The second term of Eq. (2.4) gives the spectral shape arising from the non-local exchange type excitations occurred around the core-hole site. This type of contribution is missing in the INS spectra and appears only when the theory covers \(\omega\) dependence beyond the fast collision approximation.

We end this section with the explanation how one- and two-triplon excitations are manifested in the one- and two-spin correlation functions. For simplicity, we do not mention \(n\)-triplon excitation with \(n\) more than three because \(n = 1\) and 2 dominate quantitatively. From the definitions Eqs. (2.7) and (2.8), one- and two-spin correlation functions are associated with total spin \(S = 1\) and spin-conserving excitations, respectively. Since one- and two-triplon excitations are total spin \(S = 1\) and \(S = 0, 1, 2\), respectively, one-spin correlation function includes both one- and two-triplon excitations with \(S = 1\) while two-spin correlation function includes two-triplon excitation alone.

Note that, in the undoped ladder system, multi-triplon contributions with different parity do not mix because the system is invariant with respect to reflection about the centerline of the ladder. Therefore, one- and two-triplon contributions involved in the one-spin correlation function \(Y^{(1)}(\omega; q_c, q_a, \omega)\) can be separated. That is, one- and two-triplon contributions are found in the \(q_c = \pi\) and \(q_a = 0\) modes, respectively, in which the spin excitations in leg 1 and 2 are summed up in anti-phase, and in phase, respectively.

III. NUMERICAL RESULTS

In order to evaluate \(f_\nu^{(1)}(\omega)\)'s and \(f_\nu^{(2)}(\omega)\)'s, we must prepare the eigenstates and the corresponding energies of \(H_{mag}\) and \(H'_mag\), the spin Hamiltonian in the intermediate state. In addition, the incident photon energy \(\omega_i\) should be specified, which is chosen as the peak position of the absorption coefficient. In the following, we shall describe how these preparations are made.

A. Eigenstates of \(H_{mag}\) and \(H'_{mag}\)

We consider a system consisting of 2 \(\times 8\) spins of \(S = 1/2\) with periodic boundary conditions for the initial and final states, as shown in Fig. I (b). The exchange couplings are chosen here as \(J_0 = 186\) meV, \(J_L = 124\) meV, and \(J_{cyc} = 31\) meV\(^{33,42,44}\). Representing \(H_{mag}\) by a matrix of 12870 \(\times 12870\) dimensions in the subspace of the z' component of the total spin \(S_{tot}' = 0\), we diagonalize the Hamiltonian matrix. We obtain the ground state energy as \(\epsilon_g/\langle N J_z \rangle = -0.733\). The \(H'_{mag}\) is obtained from \(H_{mag}\) by eliminating the spin at the core-hole site. Therefore \(H'_{mag}\) consists of 15 spins, and may be represented by a matrix with 6435 \(\times 6435\) dimensions in the subspace of \(S_{tot}' = \pm 1/2\).
B. Absorption coefficient

By substituting the eigenvalues and the eigenstates evaluated on finite-size cluster into Eq. (A.1) of ref. [24], we obtain the $L_{2,3}$-absorption coefficient $A_j(\omega_i)$ ($j = 3/2$ or $j = 1/2$). Figure 2 shows the calculated $A_j(\omega_i)$ as a function of the incident photon energy. The origin of photon energy is set to be very close to the Lorentzian shape. The peak position of $\epsilon_{\text{core}}$'s, $\mu_i$, is slightly shifted from $\omega_i = \epsilon_{\text{core}}$. The $\epsilon_{\text{core}}$ stands for the energy required to create a $2p$ core-hole in the multiplet $j = 1/2$ or $3/2$ of the $(3d)^{10}$ configuration. For Sr$_2$Cu$_2$O$_4$, we take $\Gamma/J_\perp = 2.4$, since a typical value of $\Gamma \sim 300$ meV for Cu $L_2$-edge and $J_\perp = 124$ meV. The calculated curve turns out to be very close to the Lorentzian shape. The peak position is slightly shifted from $\omega_i = \epsilon_{\text{core}}$, instead, it is around $\omega_i = \omega_i^0 \simeq \epsilon_{\text{core}} + 1.365J_\perp$ for $\Gamma/J_\perp = 2.4$.

C. Evaluation of the coefficients

By using the eigenvalues and eigenfunctions on a two-leg ladder of $2 \times 8$ spins, we calculate the coefficients for $\omega_i = \omega_i^0$. Table I lists the calculated values. For $f^{(1)}_\mu$, $|f^{(1)}_j|$ and $|j^{(1)}_3|$ are rather smaller than $|f^{(1)}_1|$ with $\Gamma/J_\perp = 2.4$, while they become larger with $\Gamma/J_\perp$ increasing. It implies that the effect of magnetic excitations on neighboring sites increases with decreasing value of $\Gamma$. As regards $f^{(2)}_\mu$, $|f^{(2)}_2|$, $|f^{(2)}_j| \gg |j^{(2)}_4|$ for $\Gamma/J_\perp = 1.0 \sim 2.4$. This suggests that the exchange type disturbance does not extend beyond the nearest neighbor site in both leg and rung directions.

D. RIXS spectra

We calculate the correlation functions from Eqs. (2.5) and (2.6). Figure 3 shows $Y^{(1)}(\omega_i^0; q_c, q_a, \omega)$ and $Y^{(2)}(\omega_i^0; q_c, q_a, \omega)$ numerically calculated with $\Gamma/J_\perp = 2.4$. It should be reminded that $Y^{(1)}(\omega_i^0; q_c, 0, \omega)$ and $Y^{(1)}(\omega_i^0; q_c, \pi, \omega)$ include the contributions from two- and one-triplon excitations, respectively.

In Fig. 3 (a), the energy profile of $Y^{(1)}(\omega_i^0; q_c, 0, \omega)$ is plotted for accessible values of $q_c$. The lower and upper boundaries of the two-triplon continuum, shown in Schlappa et al.'s paper are also illustrated. The peak position of $Y^{(1)}(\omega_i^0; q_c, 0, \omega)$ captures well the dispersive behavior of the lower boundary. Quantitative discrepancy can be ascribed to the finite size effect. Since Eq. (2.5) gives no finite intensity at the zone center $q_c = 0$, our theory, unfortunately, cannot observe the spin gap directly. However, a little larger cluster may give the better extrapolation of the magnitude of the spin gap at the zone center.

In Fig. 3 (b), we can see that the $q_c$ dependence of the peak position of $Y^{(1)}(\omega_i^0; q_c, \pi, \omega)$ traces one-triplon dispersion relation. Although Schlappa et al. fixed their experimental configuration such that the scattering vec-
TABLE I: Coefficients $f^{(1)}_{\nu} (\omega_0^c)$’s and $f^{(2)}_{\nu} (\omega_0^c)$’s in units of $1/J_\perp$ for different values of $\Gamma/J_\perp$ evaluated for $2 \times 8$ spin ladder.

| $\Gamma/J_\perp$ | $f^{(1)}_1 (\omega_0^c)$ | $f^{(1)}_2 (\omega_0^c)$ | $f^{(1)}_3 (\omega_0^c)$ | $f^{(1)}_4 (\omega_0^c)$ |
|-----------------|----------------------|----------------------|----------------------|----------------------|
| 2.4             | (0.034, -0.849)      | (0.052, -0.037)      | (0.029, -0.026)      | (0.035, -0.026)      |
| 1.6             | (0.063, -1.285)      | (0.084, -0.085)      | (0.045, -0.055)      | (0.056, -0.060)      |
| 1.0             | (0.019, -2.080)      | (0.110, -0.195)      | (0.056, -0.116)      | (0.073, -0.134)      |

The peak intensity of $q_c$ peaks at higher energy region as the projected momentum transfer $q_\perp$ was confined within the $bc$-plane, RIXS spectra can observe the one-triplon dispersion around the zone center $q_c \sim 0$ if the projected momentum transfer $q_\perp$ could be set as $\pi$. Such a possibility shall be discussed in Sec. IV.

The spectral shape of $Y^{(2)} (\omega_0^c; q_c, \omega)$ as a function of $\omega$ is shown in Figs. (c) and (d). It seems to have more weights at higher $\omega$ than those of $Y^{(1)} (\omega_0^c; q_c, \omega)$. This tendency is the same as observed in the analysis of one-dimensional system. The peak intensity of $Y^{(2)} (\omega_0^c; q_c, 0, \omega)$ is very small when $q_c = 0 \sim \pi/4$, but grows rapidly when $q_c$ reaches $\pi/2$. Then, the spectrum becomes broader and the spectral weight shifts to the higher energy region as $q_c$ goes to the zone boundary. The peak intensity of $Y^{(2)} (\omega_0^c; q_c, \pi, \omega)$ is small at $q_c = 0$, peaks at $q_c = \pi/2$, and vanishes at the zone boundary.

Figure 4 shows the integrated intensities defined by

$$ I^{(1)} (\omega_0^c; q_c, q_\perp) = \int Y^{(1)} (\omega; q_c, q_\perp, \omega) \frac{d\omega}{2\pi}, \quad (3.1) $$

$$ I^{(2)} (\omega_0^c; q_c, q_\perp) = \int Y^{(2)} (\omega; q_c, q_\perp, \omega) \frac{d\omega}{2\pi}. \quad (3.2) $$

FIG. 4: (Color online) Frequency-integrated intensities of the correlation functions (a) $I^{(1)} (\omega_0^c; q_c, q_\perp)/N$ and (b) $I^{(2)} (\omega_0^c; q_c, q_\perp)/N$ calculated on a $2 \times 8$ spin ladder as a function of $q_c$. (Black) circles and (red) triangles are intensities for $q_\perp = 0$ and $q_\perp = \pi$, respectively. Filled and open symbols correspond to $\Gamma/J_\perp = 2.4$ and 1.0, respectively. Solid and broken lines are guides to the eye.

FIG. 5: (Color online) A schematic diagram of the RIXS experimental configuration. The incident and scattering angles are $\theta$ and $\psi$, respectively. The scattering plane is perpendicular to the $ca$-plane. The angle between the scattering plane and the $c$-axis is $\varphi$, which is fixed to zero in the actual experiment. Scattering vector is defined as $-\mathbf{q} = \mathbf{q}_f - \mathbf{q}_i$.

The $I^{(1)} (\omega_0^c; q_c, 0)$ vanishes with $q_c \to 0$, and increases gradually with $q_c \to \pi$. The $I^{(1)} (\omega_0^c; q_c, \pi)$, on the other hand, remains finite at $q_c = 0$, and increases rapidly with $q_c \to \pi$. The presence of $I^{(2)}_2 (\omega_0^c)$ and $I^{(1)}_4 (\omega_0^c)$ makes the $q_c$-dependence deviate from that of the dynamical structure factor predicted by the fast collision approximation. The deviation is small for $\Gamma/J_\perp = 2.4$, but becomes conspicuous with $\Gamma/J_\perp = 1.0$, because of the increase of $|I^{(1)}_2 (\omega_0^c)|$ and $|I^{(1)}_4 (\omega_0^c)|$. The $I^{(2)} (\omega_0^c; q_c, 0)$ is very small but finite at $q_c = 0$, increases as $q_c$ grows peaking around $q_c = 3\pi/4$, then remains finite at the zone boundary. The $I^{(2)} (\omega_0^c; q_c, \pi)$ starts finite at $q_c = 0$, peaks around $q_c = \pi/2$, then vanishes at the zone boundary. The $I^{(2)} (\omega_0^c; q_c, \pi)$ is found one order of magnitude smaller than $I^{(1)} (\omega_0^c; q_c, q_\perp)$ over the entire Brillouin zone.

IV. APPLICATION TO Sr$_{14}$Cu$_{24}$O$_{41}$

Now we attempt to compare our results with those observed by the RIXS experiment in two-leg spin ladder system Sr$_{14}$Cu$_{24}$O$_{41}$. The legs and rungs of the lad-
The direction of the scattering vector in the experiment is performed by Schlappa. Figure 5 illustrates a schematic sketch of the experiment observed at the Cu L$_3$-edge in Sr$_2$Cu$_2$O$_{41}$. Notice that the direction of the scattering vector in the experiment is opposite to that of ours. In the following, when the finite numerical values of the momentum transfer $q_c$ and $q_a$ are mentioned, we adopt the experimental definition. The surface of the sample is perpendicular to the b-axis. Since the scattering plane is fixed parallel to the $bc(zy)$-plane, the momentum transfer along the rung direction is zero ($q_a = 0$). Hence, the experiment probed $Y^{(1)}(\omega_i; q_c, 0, \omega)$ and $Y^{(2)}(\omega_i; q_c, 0, \omega)$. The incident and scattering angles are $\theta$ and $\psi$, respectively. In the experiment, both $\psi = 90^\circ$ and $130^\circ$ were used. We choose the latter since it covers approximately $90\%$ of the Brillouin zone along the c-direction at the Cu L$_3$-edge. By changing $\theta$, the RIXS spectra for different $q_c$ were obtained. The polarization vector of the incident photon is then expressed in the $xyz$ coordinate as $\alpha_i = (0, -1, 0)$ for the $\sigma$ polarization and $\alpha_i = (\chi^\pi_i, 0, \chi^\sigma_i)$ for the $\pi$ polarization. Similarly, the polarization of the scattered photon is expressed as $\alpha_f = (0, -1, 0)$ for the $\sigma'$ polarization and $\alpha_f = (\chi^\pi_f, 0, \chi^\sigma_f)$ for the $\pi'$ polarization. The polarization is usually separated with the incident photon, but not separated with the scattered photon in experiments. In such a situation, we may express the RIXS spectra depending on the polarization of the incident photon as

$$I(\omega_i; q_c, q_a, \omega) \propto \left[ \left( \frac{\chi^\pi_i}{2} \right)^2 Y^{(1)}(\omega_i; q_c, q_a, \omega) + \left( \frac{\chi^\sigma_i}{2} \right)^2 Y^{(2)}(\omega_i; q_c, q_a, \omega) \right],$$

(4.1)

for $\sigma$-polarization, and

$$I(\omega_i; q_c, q_a, \omega) \propto \left[ \left( \frac{\chi^\pi_i}{2} \right)^2 Y^{(1)}(\omega_i; q_c, q_a, \omega) + \left( \frac{\chi^\sigma_i}{2} \right)^2 Y^{(2)}(\omega_i; q_c, q_a, \omega) \right],$$

(4.2)

for $\pi$-polarization. The contribution of $Y^{(2)}(\omega_i; q_c, q_a, \omega)$ relative to that of $Y^{(1)}(\omega_i; q_c, q_a, \omega)$ is enhanced by $(2/\chi^\pi_i)^2$ in the $\sigma$ polarization. The contribution of $Y^{(2)}(\omega_i; q_c, q_a, \omega)$ in the $\pi$ polarization is smaller than that in the $\sigma$ polarization by a factor $(\chi^\pi_i/\chi^\sigma_i)^2$.

FIG. 6: (Color online) RIXS spectra as a function of the energy loss $\omega$ for the available momentum transfer projected on the $c$-axis ($q_c$) in the $\sigma$ polarization. Solid (black) and long-dotted (red) lines show the total intensity and the contributions from $Y^{(2)}(\omega_i; q_c, q_a, \omega)$, respectively. The calculated curves are convoluted with the Lorentz function with the half-width of half-maximum 78 meV. (a) For $q_a = 0$. Filled circles are the experimental data. Thin (blue) curves are the lower and upper boundaries expected from two-triplon continua while (b) For $-q_a = \pi$. Thin (blue) line is the dispersion curve expected from one-triplon excitation.

ladders are along the $c$ and $a$ axes with lattice constants $a = 11.459\AA$, $b = 13.368\AA$, and along the $c$-direction the unit cell for the ladders is $c_L = 3.931\AA$. The lattice parameter is $a_c = 3.84\AA$ along the $a$-directions for the ladders.

A. $L_3$-edge spectrum

In this section, we compare our results with the experiment observed at the Cu $L_3$-edge in Sr$_2$Cu$_2$O$_{41}$. Figure illustrates a schematic sketch of the experimental geometry performed by Schlappa et al. Notice that the direction of the scattering vector in the experiment is opposite to that of ours. In the following, when the finite...
toward the zone boundary. In Schlappa et al. in the higher energy transfer region. The intensity grows −ev. However, around −qc ≃ −0.8π ∼ −0.6π, the tail of the intensity extends to the higher energy side about 0.2 eV. The calculated curves are convoluted with the Lorentz function with the half-width of half-maximum 78 meV. The intensity is roughly the same order of magnitude observed in the available momentum transfer projected on the c-axis (qc) in the σ polarization. Solid (black) and long-dotted (red) lines show the total intensity and the contributions from Y(2)(ωc; qc, qa, ω), respectively. The calculated curves are convoluted with the Lorentz function with the half-width of half-maximum 78 meV.

Next, we concentrate on the contribution of Y(2)(ωc; qc, qa, ω), which is smaller than that of Y(1)(ωc; qc, qa, ω) even in the presence of the geometrical coefficients appeared in Eqs. (4.1) and (4.2). Though it is small, the contribution of Y(2)(ωc; qc, qa, ω) has weight in the higher energy transfer region. The intensity grows larger as |qc| increases with |qc| → π/2, then decreases toward the zone boundary. In Schlappa et al.’s data, for each qc, the intensity is accumulated within the region centered at the peak position with half-width about 0.1 eV. However, around −qc = −0.8π ∼ −0.6π, the tail of the intensity extends to the higher energy side about 0.2 eV. This trend coincides with that shown by the two-spin correlation function, which leads us to speculate that the observed weak intensities in the high energy region may be attributed to the contribution of Y(2)(ωc; qc, qa, ω).

V. SUMMARY AND DISCUSSION

We have studied the magnetic excitation spectra of the L-edge RIXS in undoped cuprate, in particular, in quasi one-dimensional two-leg spin ladder system. We have analyzed the second-order dipole allowed process through the intermediate state, in which there is no spin degree of freedom at the core-hole site. This nature of the intermediate state is found to affect strongly the transition amplitudes of spin excitations. Then, the RIXS spectra have been derived as the one-spin and two-spin correlation functions in the channels with and without changing polarization, respectively. Note that they include the contributions of the magnetic excitations not only on the core-hole site but also on the neighboring sites. The correlation functions are expressed by a set of numerical coefficients reflecting the weight of each magnetic excitations.

FIG. 7: (Color online) RIXS spectra at the Cu M3-edge as a function of the energy loss ω for the available momentum transfer projected on the c-axis (qc) in the σ polarization. Solid (black) and long-dotted (red) lines show the total intensity and the contributions from Y(2)(ωc; qc, qa, ω), respectively. The calculated curves are convoluted with the Lorentz function with the half-width of half-maximum 78 meV.
obtained, we could concentrate on evaluating them with approximation methods available. The coefficients could be rather accurately obtained numerically in a system with small size, since the relevant excitations are restricted around the core-hole site. We have evaluated them and, subsequently, the correlation functions in a finite-size two-leg ladder. In the two-leg ladder system, the one-spin correlation function includes both one- and two-triplon excitations with $S = 1$. We find one- and two-triplon excitations emerge separately by choosing rung wave number $-q_a = \pi$ and 0, respectively. An application to Sr$_{14}$Cu$_{24}$O$_{41}$ has revealed that the calculated RIXS spectrum captures well the dispersive behavior shown by the lower boundary of two-triplon continuum. By adjusting the geometrical configuration of the experiment, one-triplon dispersion around the zone center could be detectable in the RIXS measurement. The observed weak intensity in the higher energy region around $-q_c \approx -0.87\pi - 0.67\pi$ might be the contribution from the two-spin correlation function, which could be clearly detected at the Cu $M_3$-edge.

We finally comment on the plausibility to adopt spin only model to explain the current experiment. Although our theory has reproduced semi-quantitatively well the lower boundary of the two-triplon continuum over a wide range of the Brillouin zone, the corresponding spin correlation function has no intensity at the zone center. On the other hand, a small cluster analysis on the Hubbard model presented by Schlappa et al. has given a finite intensity at $q_e = 0$. Since the RIXS processes in the actual materials are very complicated, there is a possibility that the observed RIXS signals involves something missing in the spin correlation function. For the $K$-edge RIXS spectra, for instance, Jia et al. has demonstrated that the RIXS intensity and the spin dynamical structure factor show difference in several systems. Whether the same is true to the present system is an intriguing problem and relegated to a future study.

Acknowledgments

We thank M. Grioni and H. M. Rønnow for valuable discussions. T. N. thanks to T. Hikihara for helpful discussions. This work was partially supported by a Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology of the Japanese Government.

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