SPECTRAL WEIGHT OF RESONANT INELASTIC X-RAY SCATTERING IN DOPED CUPRATES: EFFECT OF CORE-HOLE LIFETIME

TAKAMI TOHYAMA
Department of Applied Physics, Tokyo University of Science, Tokyo 125-8585, Japan
tohyama@rs.tus.ac.jp

KENJI TSUTSUI
Synchrotron Radiation Research Center, National Institutes for Quantum and Radiological Science and Technology, Hyogo 679-5148, Japan
tutui@spring8.or.jp

October 5, 2018

We examine the effect of core-hole lifetime on the spectral weight of resonant inelastic x-ray scattering (RIXS) in hole-doped cuprates. We calculate the spectral weight by using exact diagonalization technique for a $4 \times 4$ doped Hubbard lattice and find that spin-flip channel detecting single-magnon excitation is less sensitive to the core-hole lifetime while in non-spin-flip channel the spectral weight is strongly dependent on the lifetime. In the latter, charge and two-magnon excitations predominately contribute to RIXS for short and long core-hole lifetimes, respectively. For a realistic value of the core-hole lifetime in cuprates, both the charge and two-magnon excitations are expected to contribute to non-spin-flip channel in RIXS when the incident-photon energy is tuned to the main peak of x-ray absorption spectrum.

Keywords: RIXS; cuprates; Hubbard model.

1. Introduction

Resonant inelastic x-ray scattering (RIXS) experiments tuned to Cu $L$ edge have provided a lot of new insights about spin dynamics in spin-flip channel\cite{112} as well as charge dynamics in non-spin-flip channel\cite{113} in cuprate superconductors. The two channels are selected predominately by the polarization dependence of incident photon. Not only such polarization dependence but also incident-photon energy dependence of the RIXS spectrum gives us useful information on the electronic states of hole-doped cuprates. In particular, non-spin-flip channel exhibits a fluorescence-like shift of spectral weight with increasing the incident-photon energy\cite{115,110}. Such a fluorescence-like shift has been interpreted as a consequence of a change of spectral weight distribution from two-magnon excitations appearing when the incident-phonon energy is tuned to the main peak of x-ray absorption spectrum (XAS) to charge excitations of hole carriers appearing when the incident-phonon energy is...
tuned for to a satellite structure in XAS.\textsuperscript{7} The spectral weight in Cu $L$-edge RIXS is also dependent on the lifetime of core hole in the intermediate state.\textsuperscript{8} Expanding the resolvent of the intermediate state in terms of the core-hole lifetime, i.e., ultrashort core-hole lifetime expansion,\textsuperscript{9} we may apply a perturbation theory for the RIXS spectrum. If the core-hole lifetime is short enough, spectral weight in spin-flip (non-spin flip) channel is represented by the dynamical spin (charge) structure factor, which is called fast collision approximation.\textsuperscript{10} However, spectral weights have been reported to be strongly sensitive to the order of the expansion.\textsuperscript{11} This shortcoming also leads to alternative way to construct an effective low-energy operator for RIXS.\textsuperscript{12}

In this paper, we examine the effect of core-hole lifetime on RIXS spectral weight by using unbiased numerical exact diagonalization for a small two-dimensional Hubbard lattice representing doped cuprates. We find that spin-flip channel is relatively insensitive to the core-hole lifetime. In contrast, spectral weights for non-spin-flip channel depend on the value of the core-hole lifetime: charge and two-magnon excitations predominately contribute to RIXS for short and long core-hole lifetimes, respectively.

This paper is organized as follows. The Hubbard model and RIXS spectra decomposed into spin-flip and non-spin-flip channels are introduced in Sec. 2. In Sec. 3, we calculate the core-hole lifetime dependence of RIXS spectra for both spin-flip and non-spin-flip channels, and discuss their implications. A summary is given in Sec. 4.

\section{Hubbard model and RIXS spectrum}

In order to describe $3d$ electrons in the CuO$_2$ plane, we take a single-band Hubbard model given by

\begin{equation}
H_{3d} = -t \sum_{i\delta} c_{i\sigma}^\dagger c_{i+\delta\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow},
\end{equation}

where $c_{i\sigma}^\dagger$ is the creation operator of an electron with spin $\sigma$ at site $i$; number operator $n_{i\sigma} = c_{i\sigma}^\dagger c_{i\sigma}$; $i+\delta$ represents the four first nearest-neighbor sites around site $i$; and $t$ and $U$ are the nearest-neighbor hopping and on-site Coulomb interaction, respectively. Next-nearest-neighbor hoppings are neglected for simplicity.

Tuning polarization of incident and outgoing photons in RIXS, we can separate an excitation with the change of total spin by one, i.e., spin-flip channel with $\Delta S = 1$ and an excitation with no change of total spin, i.e., non-spin-flip channel with $\Delta S = 0$\textsuperscript{12,13,14,15}. The two excitations can be defined as

\begin{align}
I_{\text{RIXS}}^{\Delta S = 0}(q, \omega) &= \sum_f \left| \langle f | N^j_q | 0 \rangle \right|^2 \delta (\omega - E_f + E_0), \\
I_{\text{RIXS}}^{\Delta S = 1}(q, \omega) &= \sum_f \left| \langle f | S^j_q | 0 \rangle \right|^2 \delta (\omega - E_f + E_0),
\end{align}
with $S^j_q = (B^j_{q\uparrow\uparrow} - B^j_{q\downarrow\downarrow})/2$, $N^j_q = B^j_{q\uparrow\downarrow} + B^j_{q\downarrow\uparrow}$, and

$$B^j_{q\sigma'\sigma} = \sum_l e^{-i\mathbf{q}\cdot\mathbf{R}_l} c^\dagger_{l\sigma'} \frac{1}{\omega_l - H^j_l + E_0 + i\Gamma c^\dagger_{l\sigma}},$$

(4)

where $|0\rangle$ ($|f\rangle$) represents the ground state (the final state) with energy $E_0$ ($E_f$) in the Hubbard model (1); $j$ is the total angular momentum of Cu2p with either $j = \frac{1}{2}$ or $j = \frac{3}{2}$; $\Gamma$ is the inverse of core-hole lifetime; and $H^j_l = H_{3d} - U_c \sum_\sigma n_{l\sigma} + \varepsilon_j$ with $U_c$ and $\varepsilon_j$ being the Cu 2p-3d Coulomb interaction and energy level of Cu 2p, respectively. Here, we assume the presence of a Cu2p core hole at site $l$. $\mathbf{R}_l$ is the position vector at site $l$. $\omega_i$ in the denominator of Eq. (4) represents the incident-photon energy tuned to the main peak position of XAS calculated by

$$I^{\text{XAS}}(\omega) = -\frac{1}{\pi} \text{Im} \left[ \sum_{\omega_l} c^\dagger_{l\sigma} \frac{1}{\omega - H^j_l + E_0 + i\pi} c^\dagger_{l\sigma} |0\rangle \right].$$

(5)

When $\Gamma$ is much larger than the remaining terms in the denominator of Eq. (4), $S^j_q$ and $N^j_q$ reduce to $S^j_q = \sum_l e^{-i\mathbf{q}\cdot\mathbf{R}_l} S^j_l$ and $N_q = \sum_l e^{-i\mathbf{q}\cdot\mathbf{R}_l} N_l$, respectively, with the $z$ component of the spin operator $S^z_l$ and electron-number operator $N_l$. This is nothing but the first-collision approximation, where Eqs. (2) and (3) read the dynamical charge structure factor,

$$N(q, \omega) = \sum_f |\langle f | N_q |0\rangle|^2 \delta(\omega - E_f + E_0),$$

(6)

and the dynamical spin structure factor,

$$S(q, \omega) = \sum_f |\langle f | S^z_q |0\rangle|^2 \delta(\omega - E_f + E_0),$$

(7)

respectively.

In order to calculate Eqs. (2), (3), (5), (6), and (7), we use a Lanczos-type exact diagonalization technique on a $4 \times 4$ cluster under periodic boundary conditions. We consider hole doping with hole concentration $x = 1/8$, i.e., 2-hole doped $4 \times 4$ cluster. We take $U/t = U_c/t = 10$ and $\Gamma = t$ for XAS but $\Gamma$ in RIXS is regarded as a parameter to examine the effect of the core-hole lifetime on RIXS spectra.

3. Core-hole lifetime dependence of RIXS spectra

Before examining $\Gamma$ dependence, we calculate RIXS spectra for all of $q$ defined in the $4 \times 4$ cluster. Figure 1 shows both cases of $\Delta S = 1$ and $\Delta S = 0$ for $\Gamma = 1$. The spectra for $\Delta S = 1$ originating from single-magnon excitations are distributed lower in energy than those for $\Delta S = 0$ as expected. In particular, at $q = (\pi, \pi)$ the spectra are well separated between low-energy region for $\Delta S = 1$ and high-energy region up to $\omega = 6t$ for $\Delta S = 0$. At $q = (0, 0)$, there is no spectral weight for $\Delta S = 1$, while there appears small weight around $\omega = 0.7t$ for $\Delta S = 0$. This contrasting behavior indicates that spin-flip channel resembles to $S(q, \omega)$ whose weight at $q = (0, 0)$ is
Fig. 1. RIXS spectra at various \( q \) on the \( 4 \times 4 \) single-band Hubbard cluster with \( U = U_c = 10t \) and \( \Gamma = t \) for hole doping with hole concentration \( x = 2/18 = 0.125 \). Black and red colors represent \( \Delta S = 1 \) and \( \Delta S = 0 \) excitations, respectively. The solid curves are obtained by performing a Lorentzian broadening with a width of 0.2t. The incident photon energy is set to the main peak of absorption spectrum.

zero, while non-spin-flip channel does not necessarily represent \( N(q, \omega) \) having no weight at \( q = (0,0) \).

In order to examine \( \Gamma \) dependence, we focus on RIXS spectra at \( Q = (\pi, 0) \). Figure 2 shows the \( \Gamma \) dependence for \( \Delta S = 1 \). The spectrum at large \( \Gamma \) is the same as \( S(q, \omega) \) as expected. We find that basic spectral features of \( S(q, \omega) \) remain even for small \( \Gamma \) down to \( \Gamma = 0.1t \). This means that spin-flip channel in RIXS for cuprates predominately describes single-magnon excitations expressed by the dynamical spin structure factor.

In contrast to the spin-flip \( \Delta S = 1 \) case, non-spin-flip channel with \( \Delta S = 0 \) exhibits strong \( \Gamma \) dependence. Figure 3 shows the \( \Gamma \) dependence for \( \Delta S = 0 \). The spectrum at large \( \Gamma \) is again the same as \( N(q, \omega) \) as expected. However, with decreasing \( \Gamma \), spectral distribution deviates from \( N(q, \omega) \) with the reduction of spectral weight, and at the same time low-energy weight at \( \omega = 1.2t \) largely increases. At the smallest value of \( \Gamma \), the low-energy peak dominates spectrum. When \( \Gamma \) is comparable with the remaining terms in the denominator of Eq. (4), intersite operators emerge as effective operators in Eq. (4), in addition to the on-site charge operator \( N_i \). In non-spin-flip channel, a two-magnon-type operator is easily expected to contribute to \( N_q \) in Eq. (2). In fact, comparing with dynamical two-magnon correlation function at \( q = (\pi, 0) \) (not shown), we find that the low-energy peak corresponds to a two-magnon excitation.

For a realistic parameter of \( \Gamma(\sim t) \) for cuprates and \( \omega_i \) tuned to the main peak of XAS, a large amount of two-magnon contribution exists in addition to charge
Fig. 2. The inverse core-hole lifetime $\Gamma$ dependence of spin-flip $\Delta S = 1$ RIXS spectra at $\mathbf{q} = (\pi, 0)$ on the $4 \times 4$ single-band Hubbard cluster with $U = U_c = 10t$ for hole doping with hole concentration $x = 2/18 = 0.125$. Each color corresponds to a value of $\Gamma$ denoted in the panel except for the blue solid line representing the dynamical spin structure factor $S(\mathbf{q}, \omega)$.

Fig. 3. The inverse core-hole lifetime $\Gamma$ dependence of non-spin-flip $\Delta S = 0$ RIXS spectra at $\mathbf{q} = (\pi, 0)$ on the $4 \times 4$ single-band Hubbard cluster with $U = U_c = 10t$ for hole doping with hole concentration $x = 2/18 = 0.125$. Each color corresponds to a value of $\Gamma$ denoted in the panel except for the blue solid line representing the dynamical spin structure factor $N(\mathbf{q}, \omega)$. 
excitations. Therefore, it is important to carefully distinguish both contributions. In fact, the presence of these two components contribute to a fluorescence-like \( \omega \) dependence of non-spin-flip channel in RIXS for cuprates.

4. Summary

We have investigated the effect of core-hole lifetime on RIXS spectral weight by using unbiased numerical exact diagonalization for the \( 4 \times 4 \) two-dimensional doped Hubbard lattice. We find that spin-flip channel is less insensitive to the core-hole lifetime. In contrast, non-spin-flip channel, where both two-magnon excitation and charge excitation emerge, depends on the value of the core-hole lifetime: for short lifetime charge excitations are dominated, while for long lifetime two-magnon excitations control low-energy excitations. It is reasonable to detect two-magnon excitations for long core-hole lifetime, since an electron excited to the Hubbard band from core level can propagate the system during the core-hole lifetime, leading to effective intersite excitations like two magnons. We should be careful for the fact that both charge and two-magnon excitations are expected to contribute to RIXS spectra for a realistic value of the core-hole lifetime for cuprates.

Acknowledgements

This work was supported by the Japan Society for the Promotion of Science, KAKENHI (Grants No. 26287079, No. 15H03553, and No. 16H04004) and Creation of new functional devices and high-performance materials to support next-generation industries (GCDMSI) to be tackled by using post-K computer and by MEXT HPCI Strategic Programs for Innovative Research (SPIRE) (hp160222, hp170274) from Ministry of Education, Culture, Sports, Science, and Technology.

1. L. J. P. Ament, M. van Veenendaal, T. P. Devereaux, J. P. Hill, and J. van den Brink, Rev. Mod. Phys. 83, 705 (2011).
2. For a review, M. P. M. Dean, J. Magn. Magn. Mater. 376, 3 (2015) and references therein.
3. K. Ishii, M. Fujita, T. Sasaki, M. Minola, G. Dellea, C. Mazzoli, K. Kummer, G. Ghiringhelli, L. Braicovich, T. Tohyama, K. Tsutsumi, K. Sato, R. Kajimoto, K. Ikeuchi, K. Yamada, M. Yoshida, M. Kurooka and J. Mizuki, Nat. Commun. 5, 3714 (2014).
4. M. Guarise, B. Dalla Piazza, H. Berger, E. Giannini, T. Schmitt, H. M. Rennow, G.A. Sawatzky, J. van den Brink, D. Altenfeld, I. Eremin, and M. Grioni, Nat. Commun. 5, 5760 (2014).
5. M. Minola, G. Dellea, H. Gretarsson, Y. Y. Peng, Y. Lu, J. Porras, T. Loew, F. Yakhou, N. B. Brookes, Y. B. Huang, J. Pelliciari, T. Schmitt, G. Ghiringhelli, B. Keimer, L. Braicovich, and M. Le Tacon, Phys. Rev. Lett. 114, 217003 (2015).
6. H. Y. Huang, C. J. Jia, Z. Y. Chen, K. Wohlfeld, B. Moritz, T. P. Devereaux, W. B. Wu, J. Okamoto, W. S. Lee, M. Hashimoto, Y. He, Z. X. Shen, Y. Yoshida, H. Eisaki, C. Y. Mou, C. T. Chen, and D. J. Huang, Sci. Rep. 6, 19657 (2016).
7. K. Tsutsui and T. Tohyama, Phys. Rev. B 94, 085144 (2016).
8. J. van den Brink and M. van Veenendaal, Europhys. Lett. 73, 121 (2006).
9. L. J. P. Ament, F. Forte, and J. van den Brink, Phys. Rev. B 75, 115118 (2007).
10. L. J. P. Ament, G. Ghiringhelli, M. M. Sala, L. Braicovich, and J. van den Brink, *Phys. Rev. Lett.* **103**, 117003 (2009).
11. C. Jia, K. Wohlfeld, Y. Wang, B. Moritz, and T. P. Devereaux, *Phys. Rev. X* **6**, 021020 (2016).
12. M. W. Haverkort, *Phys. Rev. Lett.* **105**, 167404 (2010).
13. J. I. Igarashi and T. Nagao, *Phys. Rev. B* **85**, 064421 (2012).
14. S. Kourtis, J. van den Brink, and M. Daghofer, *Phys. Rev. B* **85**, 064423 (2012).
15. T. Tohyama, K. Tsutsui, M. Mori, S. Sota, and S. Yunoki, *Phys. Rev. B* **92**, 014515 (2015).