Theoretical study of orbital excitations by resonant inelastic x-ray scattering in doped manganites

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Abstract

Resonant inelastic x-ray scattering in doped ferromagnetic manganites with orbital disorder is examined by using the exact diagonalisation method in finite size cluster systems. The calculated spectra of the dynamical charge correlation functions consist of the intra-band and inter-band excitations. The intensity corresponding to the intra-band excitations increases with doping of holes. This feature is consistent with the recent experiments of the resonant inelastic x-ray scattering in $La_{1-x}Sr_xMnO_3$ (x=0.2, and 0.4).

Key words: orbital ordering; manganites; resonant inelastic x-ray scattering

1. Introduction

Perovskite manganites $A_{1-x}B_x\mathrm{MnO}_3$ (A=La, Pr, B=Sr, Ca) are widely accepted as a system where the orbital degree of freedom plays essential roles in various magnetic, optical and transport properties. Orbital ordering in manganites has been studied by resonant x-ray scattering [1]. By extending the studies, the resonant inelastic x-ray scattering (RIXS) is used to examine excitations in the orbital ordered states. The excitations in the orbital ordered $LaMnO_3$ where $d_{3x^2-r^2}$ and $d_{3y^2-r^2}$ orbitals align alternately in the xy plane have been recently studied by RIXS experimentally [2] and theoretically [3]. It was clarified that a newly observed peak around 2.5 eV in the RIXS experiments is attributed to the excitations from the effective lower-Hubbard band with $d_{3x^2-r^2}$ and $d_{3y^2-r^2}$ orbitals to the upper-Hubbard one with $d_{x^2-z^2}$ and $d_{z^2-y^2}$ orbitals. On the other hand, the excitations in the orbital disordered state in manganites are still unknown. Recently, the RIXS experiments in ferromagnetic $La_{1-x}Sr_xMnO_3$ (x=0.2, and 0.4), where the orbital is considered to be disordered, have been done [4]. It is shown that the RIXS intensity below 2 eV is enhanced with doping of holes.

In this paper, we study the RIXS spectra in the doped ferromagnetic manganites with orbital disorder. In the studies of the RIXS spectra in cuprates, it is confirmed that the nature of RIXS spectra in the intra-band excitations are well reproduced by the dynamical charge correlation functions [5]. Therefore instead of the RIXS spectra, we calculate the dynamical charge correlation functions and focus on the intra-band excitations. We adopt the exact diagonalisation method in finite size cluster systems. The calculated spectra are consistent with the RIXS experimental results [4].

2. Numerical results

In order to study RIXS in ferromagnetic manganites we consider the Hamiltonian given by

$$H_{3d} = \sum_{<l,l'>\gamma,\gamma'} (t_{l,l'}^{\gamma;\gamma'} d_{l,\gamma}^{\dagger} d_{l',\gamma'} + \mathrm{H.c.}) + U \sum_{l,\gamma} n_{l,\gamma} n_{l,\overline{\gamma}},$$

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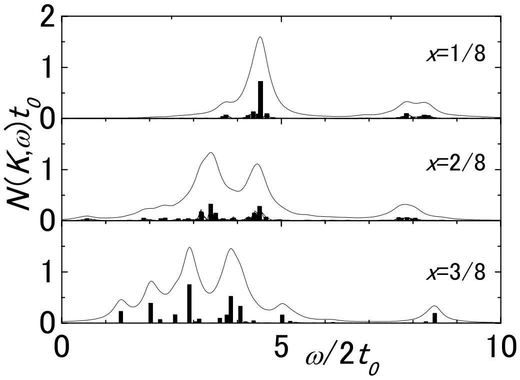

Fig. 1. The dynamical charge correlation functions at the hole concentration x=1/8, 2/8, and 3/8. The momentum transfer and the inter-orbital Coulomb interaction are chosen to be K=(π,π,π) and U/t_0=12, respectively. The spectra above and below $\omega/2t_0$=$6t_0$ are attributed to the inter-band and intra-band excitations, respectively.

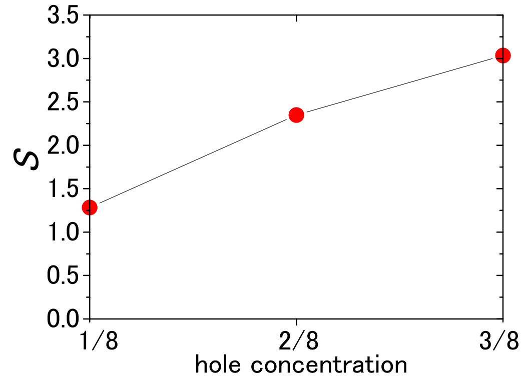

Fig. 2. The integrated intensity of $N(K,\omega)$ defined by $S=\int_0^{12t_0} d\omega N(K,\omega)$ at K=(π,π,π). The hole concentrations are chosen to be x=1/8, 2/8, and 3/8.

where $d_{l,\gamma}$ ($d^\dagger_{l,\gamma}$) is the annihilation (creation) operator of a $3d$ e_g electron at site l with orbital γ (=$3z^2$-r^2, x^2-y^2). The spin index is omitted because the system is in the ferromagnetic state. $t^{\gamma,\gamma'}_{l,l'}$ is the hopping integral between site l with orbital γ and l' with γ' and it is determined by the Slater-Koster parameters [6]. U is the inter-orbital Coulomb interaction and t_0 is the hopping integral between $3d_{3z^2-r^2}$ orbitals in the z direction. $n_{l,\gamma}$ is the number operator defined by $n_{l,\gamma}$=$d^\dagger_{l,\gamma}d_{l,\gamma}$ and $\overline{\gamma}$ indicates the counter part of γ. We calculate the charge dynamical correlation functions by using the exact diagonalisation method in finite size cluster systems. The dynamical charge correlation function is defined by

$$N(K,\omega)=\sum_f |\langle f|\, n_K\, |i\rangle|^2\, \delta(\varepsilon_f-\varepsilon_i-\omega),$$

with $n_K=\sum_{k,\gamma} d^\dagger_{k+K,\gamma}d_{k,\gamma}$. $|i\rangle$ indicates a ground state with energy ε_i and $|f\rangle$ indicates an excited one with ε_f. We choose a three dimensional cubic cluster consisting of 2×2×2 sizes.

The calculated spectra shown in Fig. 1 consist of the intra-band excitations below $12t_0$ and inter-band excitations above $12t_0$. We focus on the intra-band excitations in the spectra. In Fig. 2, the integrated intensity of $N(K,\omega)$ defined by $\int_0^{12t_0} d\omega N(K,\omega)$ at K=(π,π,π) is presented. This value increases with doping of holes. The experimental RIXS spectra in $La_{1-x}Sr_xMnO_3$ (x=0.2, and 0.4) measured in SPring-8 show that the intensity of the spectra below 2 eV increases with doping of holes [4]. This feature is consistent with the presented theoretical results.

In conclusion, we have calculated the dynamical charge correlation functions in doped ferromagnetic manganites with orbital disorder by using the exact diagonalisation method. The spectral weight attributed to the intra-band excitations is enhanced with doping of holes. This feature is consistent with the recent RIXS experiments in $La_{1-x}Sr_xMnO_3$ (x=0.2, and 0.4).

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References

[1] Y. Murakami, H. Kawada, H. Kawata, M. Tanaka, T. Arima, Y. Moritomo, and Y. Tokura, Phys. Rev. Lett. **80**, (1998) 1932.

[2] T. Inami, S. Ishihara, H. Kondo, J. Mizuki, T. Fukuda, S. Maekawa, H. Nakao, T. Matsumura, K. Hirota, Y. Murakami, Y. Endoh, cond-mat/0109509.

[3] H. Kondo, S. Ishihara, and S. Maekawa, Phys. Rev. B. **64**, (2001) 014414.

[4] K. Ishii et al, to be published.

[5] K. Tsutsui et al, to be published.

[6] J. C. Slater and G. F. Koster, Phys. Rev. **94**, (1954) 1498.