

# Resonant two-magnon Raman scattering in two-dimensional Mott insulators

Hiroaki Onodera <sup>a,1</sup>, Kenji Tsutsui <sup>a</sup>, Takami Tohyama <sup>a</sup>, Sadamichi Maekawa <sup>a</sup>

<sup>a</sup>*Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan*

---

## Abstract

We investigate the resonant two-magnon Raman scattering in two-dimensional Mott insulators by using a half-filled extended Hubbard model with the second and third nearest-neighbour hopping terms,  $t$  and  $t''$ , and the nearest-neighbour Coulomb interaction,  $V$ . By performing numerical diagonalization calculations for a small cluster, we find that the model can reproduce the experimental features that the Raman intensity is enhanced when the incoming photon energy  $\omega_i$  is not near the absorption edge but well above it. We also find that the  $t$ ,  $t''$ , and  $V$  terms bring about less effect on resonant behaviours in the Raman scattering.

*Key words:* resonant two-magnon Raman scattering; Hubbard model; Mott insulators

---

In Mott insulators, low-energy physics associated with spin degree of freedom is described two-dimensional (2D) Heisenberg model. However, the nature of high-energy excited states across the Mott gap is less clear as compared with that of the low-energy spin states. Such excited states are usually investigated by optical absorption measurements, where photons excite the system across the Mott gap. In addition, resonant Raman scattering is very useful for the investigation of the photoexcited states, because the resonance is observed when incoming photon energy  $\omega_i$  is near the energy of the photoexcited states. In the Raman scattering of 2D insulating cuprates, a two-magnon peak has been observed in the  $B_1$  geometry. The  $\omega_i$  dependence of the two-magnon peak intensity shows an interesting feature that the intensity is not enhanced near the gap edge ( $\sim 1.8\text{-}2\text{eV}$ ), but resonance occurs at higher  $\omega_i$  ( $\sim 3\text{eV}$ ) [1,2].

In this paper, numerical calculations of the two-magnon Raman intensity as a function of  $\omega_i$  are performed for a half-filled 2D Hubbard model with the second and third nearest-neighbour (NN) hopping terms,  $t'$  and  $t''$ , and the NN Coulomb interaction

$V$ , in addition to the NN hopping  $t$  and the on-site Coulomb interaction  $U$ . We find that the effect of  $t'$ ,  $t''$ , and  $V$  on resonant behaviours in the Raman scattering is small and the model can reproduce the experimental observations mentioned above.

In 2D cuprates, the value of  $t$  is estimated to be  $t \sim 0.35\text{eV}$  from the analysis of the electronic structures [3]. The value of  $U$  is evaluated to be  $U = 10t$  for the gap values to be consistent with experimental ones. The long-range hoppings  $t'$  and  $t''$  are known to be necessary for the precise description of the electronic states in the cuprates and their values are determined from the analysis of angle-resolved photoemission data [3]:  $t' = -0.343t$  and  $t'' = 0.229t$ .  $V/t$  is taken to be 1.5, which is used in one-dimensional cuprates [3].

We consider the strong coupling limit ( $U \gg t$ ) of the extended Hubbard model. In this limit, the ground state and final states of the two-magnon Raman scattering have no doubly occupied site. We perform the second order perturbation of  $t$ , neglecting  $t'$  and  $t''$  in the perturbation because of small values of  $|t'|$  and  $|t''|$  as compared with  $t$ . Then, the ground and final states are described by a 2D Heisenberg model that contains only the NN exchange interaction. In the intermediate states of the Raman process, there are one doubly oc-

---

<sup>1</sup> E-mail: onoderah@imr.tohoku.ac.jp

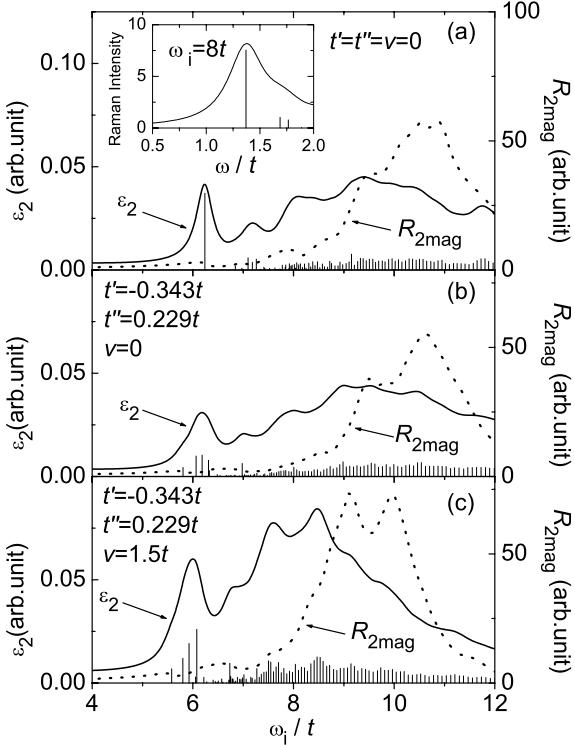


Fig. 1. The  $\omega_i$  dependence of Raman intensity  $R_{2\text{mag}}$  (dotted line), and absorption spectrum  $\varepsilon_2$  (solid line) in a 20-site cluster with  $U=10t$ , (a)  $t=t''=V=0$ , (b)  $t' = -0.343t, t'' = 0.229t, V = 0$ , (c)  $t' = -0.343t, t'' = 0.229t, V = 1.5t$ . Inset in (a) shows two-magnon Raman spectrum at  $\omega_i=8t$ . The solid lines are obtained by performing a Lorentzian broadening with of 0.4t on delta functions denoted by thin vertical bars.

cupied site and one vacant site. The expression of an effective Hamiltonian for the states within the second order perturbation of  $t$  has been given in [4]. In the present study, the  $t'$  and  $t''$  terms for the doubly occupied and vacant sites and an attractive Coulomb interaction  $-V$  between the two sites are added to the effective Hamiltonian. We calculate numerically two-magnon Raman intensity and absorption spectrum  $\varepsilon_2$  (their expressions are shown in [5]), by using numerical diagonalization method for a  $\sqrt{20} \times \sqrt{20}$  square cluster with periodic boundary condition.

First we show results for  $t=t''=V=0$  [5]. In Fig. 1(a) the inset shows two-magnon Raman spectrum at  $\omega_i=8t$ . We define  $R_{2\text{mag}}$  as the intensity of the lowest-energy peak at  $\omega=1.37t$ . The main panel of (a) shows the  $\omega_i$  dependence of  $R_{2\text{mag}}$  (dotted line) and  $\varepsilon_2$  (solid line).  $\varepsilon_2$  has an edge peak at  $\omega_i=6.2t$ . However,  $R_{2\text{mag}}$  is not enhanced at the absorption-edge peak, but the resonance occurs at  $\omega_i \sim 11t$ . This  $\omega_i$  dependence of  $R_{2\text{mag}}$  is similar to the experimental data [1,2].

Next we introduce the long-range hoppings

$t'=-0.343t$  and  $t''=0.229t$ , and set  $V=0$ . Fig. 1(b) shows the  $\omega_i$  dependence of  $R_{2\text{mag}}$  and  $\varepsilon_2$ . The tendency of the  $\omega_i$  dependence of  $R_{2\text{mag}}$  is the same as that in Fig. 1(a).

Finally we introduce both the long-range hopping and NN Coulomb interaction  $V=1.5t$ . Fig. 1(c) shows the  $\omega_i$  dependence of  $R_{2\text{mag}}$  and  $\varepsilon_2$ . The tendency of the  $\omega_i$  dependence of  $R_{2\text{mag}}$  is similar to the previous two cases. We note that the magnitude of  $R_{2\text{mag}}$  and  $\varepsilon_2$  is enhanced as compared with that in Fig. 1(b). This is related to the feature that  $V$  arranges the doubly occupied and vacant sites with one lattice spacing. We also note that the energy difference between absorption-edge and resonance maximum is about  $4t \sim 1.4\text{eV}$ , being consistent with experimental data of  $1.35\text{eV}$  [2].

In our previous study, we investigated the photoexcited states of the Hubbard model without  $t', t'', V$  [5]. In the study we found that the photoexcited states of absorption-edge are characterised by the spin degree of freedom and the photoexcited states for the resonance of two-magnon Raman intensity are characterised by the charge degree of freedom.

In summary, we have calculated resonant two-magnon Raman intensity with the extended Hubbard model in the strong coupling limit. The  $\omega_i$  dependence of Raman intensity is qualitatively unchanged by inclusion of  $t', t''$  and  $V$ . The dependence is in agreement with experiments.

This work was supported by a Grant-in-Aid for scientific Research from the Ministry of Education, Culture, Sports, Science and Technology of Japan, and CREST. One of the authors (S. M.) acknowledges support of the Humboldt Foundation. The numerical calculations were performed in the supercomputing facilities in ISSP, University of Tokyo, and IMR, Tohoku University.

## References

- [1] M. Yoshida, S. Tajima, N. Koshizuka, S. Tanaka, S. Utida, T. Itoh, Phys. Rev. B **46** (1992) 6505.
- [2] G. Blumberg, P. Abbamonte, M. V. Klein, W. C. Lee, D. M. Ginsberg, L. L. Miller, A. Zibold, Phys. Rev. B **53** (1996) R11930.
- [3] S. Maekawa and T. Thoyama, Rep. Prog. Phys. **64** (2001) 383; and references therein.
- [4] M. Takahashi, T. Thoyama, S. Maekawa, cond-mat/0201190.
- [5] T. Thoyama, H. Onodera, K. Tsutsui, S. Maekawa, cond-mat/0205388.