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Optical properties in 1D and 2D insulating cuprates

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The charge gap in insulating cuprates is a consequence of strong electron correlation. This is completely different from the band insulators, where the charge gap is basically originated from band effects. The nature of charge excitation across the gap is thus essentially different between the two types of insulators. Recently, anomalously enhanced third-order nonlinear optical susceptibility $\chi^{(3)}$ has been reported for one-dimensional (1D) Mott insulators [1,2]. In two-dimensional (2D) insulating cuprates, unusual features in the optical properties have been found in the dependence of the two-magnon Raman scattering intensity on the incoming photon frequency [3]. The intensity is not enhanced when the photon frequency approached the low-energy strong absorption peak but is resonantly enhanced when it becomes close to the high-energy weak absorption peak.

Motivated by these striking experiments, we theoretically examine photoexcited states in the 1D and 2D Mott insulators described by the half-filled Hubbard model. First, nonlinear optical response is investigated [4]. The $\chi^{(3)}$ is calculated by using the numerically exact diagonalization method on finite-size clusters. We find that $\chi^{(3)}$ increases with decreasing dimensionality. It is shown that in the 1D system dipole-allowed (odd) and -forbidden (even) states are almost degenerate in energy, having very large dipole coupling between the two states. We demonstrate that this peculiar feature is due to the spin-charge separation inherent in the 1D Mott insulators. Based on the spin-charge separation, we propose an effective model that can describe the optical nonlinearity, i.e., a holon-doublon model, where holon and doublon represent the charge degree of freedom for photoinduced unoccupied and doubly occupied sites, respectively. We note that the model is different from the standard exciton model of 1D semiconductors, since a holon and a doublon cannot occupy the same site due to the hard core constraint. The model reproduces very well the characteristic behaviors of the experimental $\chi^{(3)}$ in Sr_2CuO_3 and Ca_2CuO_3 [5]. Second, we examine the resonant two-magnon Raman scattering in the half-filled 2D Hubbard model. It is found that the Raman intensity is resonantly enhanced when the incoming photon frequency is close to a weak absorption structure located $\sim$1 eV above the strongest absorption peak. This is consistent with the experiments. The difference in the dipole transitions to the intermediate state from the initial and final states is the origin of such a behavior.

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