Theoretical study of nonlinear optical response in Mott insulators(Abstracts of Doctoral Dissertations,Annual Report(from April 2002 to March 2003))

Theoretical study of nonlinear optical response in Mott insulators

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Nonlinear optical response gives rich information on photoexcited states in solids. Recently, anomalously enhanced third-order nonlinear optical susceptibility ($\chi^{(3)}$) has been reported for the one-dimensional (1D) Mott insulators [1]. These experiments uncovered novel optical phenomena in Mott insulators. Nonlinear optical spectroscopy is also applied to two-dimensional (2D) Mott insulators [2-4], though the magnitude of the third-order optical susceptibility is one order smaller than that for 1D. The concept of spin-charge separation is a good starting point to describe the photoexcited states and nonlinear response in the 1D Mott insulators [5]. However, the concept dose not always work in 2D, since spin and charge degrees of freedom are coupled each other.

I theoretically examine material and dimensionality dependence of linear and nonlinear optical response in the insulating cuprates. First, I examined material dependence for nonlinear optical spectrum, using multi-band Hubbard model in which $3d$ orbital of copper and $2p$ orbital of oxygen are taken into account. Second, I introduced an effective model, in order to examine the dimensionality dependence in the insulating cuprates. The photoexcited states of the Mott insulators can be described by the effective model that is derived by mapping the multi-band Hubbard model onto the single-band one and by taking the strong-coupling limit. The Hilbert space of the effective Hamiltonian is restricted to the subspace with an empty and a doubly occupied sites. By using numerical technique for small clusters described by the two models, I examined photoexcited states and nonlinear optical response in Mott insulating cuprates and compared the theoretical results with experimental ones.

In order to understand material dependence for $\chi^{(3)}$ spectra between Sr_2CuO_3 and Ca_2CuO_3, I examined nonlinear optical susceptibility using multi-band Hubbard model. These two materials share common crystal structure. But $\chi^{(3)}$ spectra of Sr_2CuO_3 are about two times larger than those of Ca_2CuO_3. I found that calculated $\chi^{(3)}$ spectra reproduce the material dependence. The optical gap is found to be most important for material dependence of nonlinear susceptibility. Therefore, if one could make new insulating cuprates having smaller optical gap, the materials should have larger third order susceptibility. These features do not depend on dimensionality.

By examining linear absorption, two-photon absorption (TPA) and third-harmonic generation (THG) using the effective model, I clarifed the nature of photoexcited states such as the

distribution of dipole-allowed states with odd parity and dipole-forbidden states with even parity. I obtained characteristic features that are consistent with the experimental data of linear absorption and TPA for the insulating cuprates. They are (i) linear absorption spectrum in 2D has two broad peaks in contrast to a single peak in 1D, (ii) the magnitude of the TPA (THG) spectrum in 2D is smaller than that in 1D, and (iii) the peak and edge positions of the TPA spectrum in 2D are lower than those of linear absorption in contrast to the 1D where TPA and linear absorption show almost an identical behavior near the edge. The separation of charge and spin degree of freedom plays an essential role in the 1D Mott insulator, while in 2D the charge motion is strongly influenced by the presence of spins in the background. As expected from this picture, I found that the features (i) and (iii) are sensitive to the value of the exchange interaction between spins. In the THG spectrum of the 2D Mott insulators, dominant contributions are found to come from the process of three-photon resonance associated with odd-parity states. The two-photon resonance process is hidden by the dominant contributions. The spectrum obtained by using realistic parameters for 2D insulating cuprates has good resemblance to experimental THG spectra with a broad maximum [4].

By examining co-polarized and cross-polarized configurations, we are able to extract information about all symmetry of photoexcited states. In 2D system, TPA and THG spectra are calculated with both co-polarized and cross-polarized configuration and are compared with existing experimental data of $\chi^{(3)}$. It is shown in both the TPA and THG spectra that the spectral distribution is similar to $\chi^{(3)}$ with co-polarized configuration but the weight is small. In order to obtain further insight into the photoexcited states of the 2D Mott insulators, it is desired to measure experimentally the weight and distribution of the cross-polarized TPA and THG and compare them with the present results.

References

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