# Nonlinear Optical Response in two-dimensional Mott Insulators

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**Abstract**

We theoritically examine the nonlinear optical responses in two-dimensional Mott insulators. The photoexcited states of the Mott insulators are described by an effective model in the strong-coupling limit of a half-filled Hubbard model. By comparing the linear optical absorption, nonlinear optical two-photon absorption and third harmonic generation (THG), we clarify the nature of photoexcited states such as the distribution of odd-parity and even-parity states. In the THG spectrum, main contribution is found to come from the process of three-photon resonance associated with the odd-parity states. As a result, the two-photon resonance process is less pronounced in the THG spectrum. The calculated THG spectrum is qualitatively consistent with recent experimental data.

*Key words:* Optical materials ; Mott insulators ; Nonlinear optical response ; Hubbard model

The charge gap in Mott insulators is a consequence of strong electron correlation. The optical response is used to investigate the charge excitation across the Mott gap. Particularly, the nonlinear susceptibility with respect to the applied electric field, $\chi^{(3)}$, provides useful information on photoexcited states with odd parity and even parity. Recently, $\chi^{(3)}$ has been measured for one-dimensional (1D) Mott insulators of Cu oxides [1,2] and Ni halides [1]. In the studies, large values of $\chi^{(3)}(-\omega;0,0,\omega)$ and $\chi^{(3)}(-\omega_1;-\omega_2,\omega_2,\omega_1)$ have been obtained from the electro-refrectance measurements and the pump and probe spectroscopy, respectively. Here, $\chi^{(3)}$ is defined as $P(\omega_\sigma) = \chi^{(3)}(-\omega_\sigma;\omega_1,\omega_2,\omega_3)E(\omega_1)E(\omega_2)E(\omega_3)$, $P(\omega_\sigma)$ and $E(\omega)$ being nonlinear polarization with $\omega_\sigma = \omega_1+\omega_2+\omega_3$ and the electric field of light, respectively. The analysis of $\chi^{(3)}(-\omega;0,0,\omega)$ based on a three-level model has suggested that odd- and even-parity states are nearly degenerate with a large transition dipole moment between them [1]. This is consistent with the analysis of the two-photon absorption (TPA) spectrum, $\mathrm{Im}\chi^{(3)}(\omega_1;-\omega_2,\omega_2,\omega_1)$ [2].

The single-band Hubbard Hamiltonian is given by

$$H = -t\sum_{\langle i,j\rangle,\sigma}\left(c^\dagger_{i,\sigma}c_{j,\sigma}+\mathrm{H.c.}\right)+U\sum_i n_{i,\uparrow}n_{i,\downarrow} +V\sum_{\langle i,j\rangle}n_i n_j, \qquad (1)$$

where $c^\dagger_{i,\sigma}$ is the creation operator of an electron with spin $\sigma$ at site $i$, $n_i{=}n_{i,\uparrow}{+}n_{i,\downarrow}$, $\langle i,j\rangle$ runs over pairs on nearest neighbor (NN) sites, $t$ is the hopping integral, $U$ is the on-site Coulomb interaction, and $V$ is the Coulomb interaction between NN sites. In the strong coupling limit ($U >> t$), the ground state at half filling has one spin per site, i.e., there is no doubly occupied site. However, the photoexcited states that are created by the light have both one doubly occupied site and one vacant site. In order to obtain an effective Hamiltonian for the Hubbard model to describe the photoexcited states, we introduce projection operators $\Pi_0$, $\Pi_1$, and $\Pi_2$ onto the Hilbert space with no doubly occupied site, one doubly occupied site, and two doubly occupied sites, respectively. The effective Hamiltonian up to the second order of the hopping term $H_t$ in Eq.(1), is given by

$$H_{\mathrm{eff}} = \Pi_1 H_t\Pi_1 - \frac{1}{U}\Pi_1 H_t\Pi_2 H_t\Pi_1 + \frac{1}{U}\Pi_1 H_t\Pi_0 H_t\Pi_1 -V\sum_{i,j}n_{i\uparrow}n_{i\downarrow}(1-n_{j\uparrow})(1-n_{j\downarrow}) + U. \qquad (2)$$

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A complete expression of Eq. (2) has been given elsewhere [3].

In this paper, we show spectra obtained by a $4 \times 4$ site two-dimensional (2D) cluster with open boundary condition. A $4 \times 4$ site has 90-degree rotational symmetry. The ground state of this cluster is obtained by applying the Lanczos method to the Heisenberg Hamiltonian. $\chi^{(1)}$ and $\chi^{(3)}$ spectra are calculated by using the correction vector technique [3].

The pump and probe spectroscopy is employed to detect even-parity states [2,4]. In the linear absorption, odd-parity states are detected. Fig. 1(a) shows TPA $\mathrm{Im}\chi^{(3)}(-\omega; -\omega, \omega, \omega)$ (solid line) together with linear absorption $\chi^{(1)}(-2\omega; 2\omega)$ (broken line). In the figure, the two spetra are plotted with making the largest weight the same height. We find a peak position is lower in TPA (solid arrow) than in linear absorption (broken arrow of lower energy side), while in 1D a peak position is nearly identical between linear absorption and TPA spectra [3,5].

Fig. 1(b) shows THG spectrum $|\chi^{(3)}(-3\omega; \omega, \omega, \omega)|$ which contains information about both odd- and even-parity states via multi-photon resonance process. The spectrum consists of a broad peak and a hump below $\omega \sim 3.4t$. Since the peak position $\omega \sim 2.4t$ coincides with the one-third of the first-peak energy in the linear absorption spectrum ($\sim 7t$) in Fig. 1. (a), the origin of the peak can be attributed to three-photon resonance resonating with odd-parity states. On the other hand, there are two possiblitis as the origin of the broad structure around $\omega \sim 3t$. One is again three-photon resonance, and the other is two-photon resonance resonating with even-parity states. The broken arrow at $\omega = 2.9t$ in Fig. 1(b) indicates the one-third of the second-peak energy in the linear absorption spectrum ($8.7t$) of Fig. 1(a) indicated by broken arrow, while the solid arrow at $\omega = 3.2t$ indicates the energy of the lowest-energy peak in the TPA spectrum of Fig. 1(a) indicated by a solid arrow. Since the hump position is closer to the broken arrow, the hump is caused by the three-photon resonance. A contribution of the even-parity states through two-photon resonance is supposed to overlap with three-photon resonance and thus may be hidden as a background in the THG spectrum.

In summary, we have examined the photoexcited states and nonlinear optical responses in the 2D Mott insulators by using an effective model for a sigle-band Hubbard model in the strong-couple limit. We find a peak position is lower in TPA than in linear absorption. In THG spectrum, dominant contribution are found to come from the process of three-photon resonance. The two-photon renonance process is hidden by the dominant contribution. The THG spectrum shows broad spectral features, which are qualitatively in agreement with the experimental data with broad maximum [6,7].

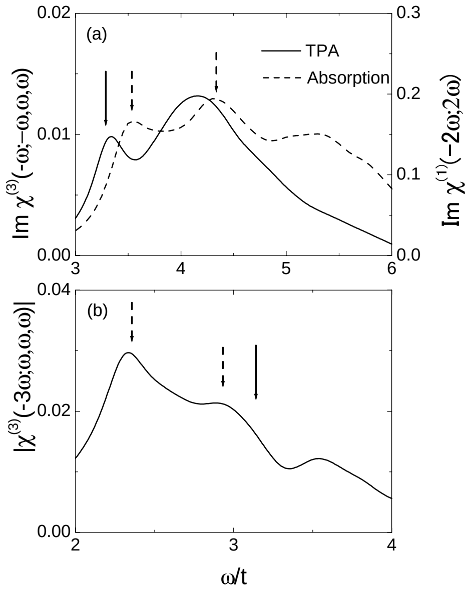


Fig. 1. (a) $\mathrm{Im}\chi^{(3)}(-\omega; -\omega, \omega, \omega)$ (solid line) and $\mathrm{Im}\chi^{(1)}(-2\omega; 2\omega)$ of an effective model with $U/t = 10$ and $V/t = 1$. The solid and broken arrows denote the peak position of TPA and $\mathrm{Im}\chi^{(1)}(-2\omega; 2\omega)$, respectively. (b) $|\chi^{(3)}(-3\omega; \omega, \omega, \omega)|$ with same parameter. The solid arrow denotes the position of two-photon resonance expected form TPA. The broken arrows denote the position of three-photon resonance expected from $\mathrm{Im}\chi^{(1)}(-2\omega; 2\omega)$.

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