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Nonlinear electrostatic response of strongly correlated one-dimensional electron systems

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Abstract

We investigate the nonlinear response of the strongly correlated one-dimensional electron systems to the static electric field with using the one-dimensional Hubbard model in the half-filled case. We adopt the variational Monte Carlo method with the Gutzwiller wave function to describe the strong correlation effects. In the weak correlation region $U/t \leq 4$, where U is the one-site Coulomb repulsion energy and t is the transfer integral between the nearest neighbor sites, the response can be described within the band picture, and the third order nonlinear susceptibility $\chi^{(3)}$ increases slowly with increasing U/t. For $U/t \leq 4$, $\chi^{(3)}$ increases rapidly with increasing U/t, and $\chi^{(3)}$ at $U/t = 10$ is more than ten times larger than that at $U/t = 2$. This large value of $\chi^{(3)}$ originates from the exotic properties of carriers in the strongly correlated one-dimensional electron systems.

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Recently, quite large third order nonlinear optical effects are observed in various strongly correlated one-dimensional electron systems (SC1DES) [1–4]. These materials have been attracting much attention because large optical nonlinearity is an important property for optical devices in future. The exact calculation study in the small size clusters showed that the large third order optical susceptibilities originate from the exotic properties of photogenerated carries in the SC1DES [5,6]. However, it is shown in the models for conjugated polymers that the magnitudes of the third order optical susceptibilities strongly depend on the system size N. That is the third order optical susceptibilities increase with increasing N with the power larger than one when N is small, and they become proportional to N when N is large enough [7–9]. Therefore, it is important to adopt the system sizes in the proportional region to consider the nonlinear optics in the thermo dynamic limit. Furthermore, it is important to consider how optical nonlinearity changes as the magnitude of Coulomb interaction is changed, to investigate the role of the correlation effect. Considering these points, we investigate the nonlinear response of the one-dimensional electron systems to the static electric field for the wide range of the correlation strength with using the large enough system size. In order to clarify the large optical nonlinearity in SC1DES, it is important to investigate the nonlinear electrostatic response, because experimental data for electrore-flectance spectra indicates that the electrostatic field significantly contributes to the optical processes in SC1DES [1]. It is also noted that in general the electrostatic response is closely correlated to the off-resonant nonlinearlity.

We consider the following Hamiltonian describing the SC1DES coupled with electrostatic field:

$$H = -t \sum_{\langle i,j \rangle,\sigma} (c_{i,\sigma}^{\dagger} c_{j,\sigma} + c_{j,\sigma}^{\dagger} c_{i,\sigma}) + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow} - E\hat{P} \qquad (1)$$

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We adopt the Hubbard Hamiltonian for SC1DES, where t is the magnitude of the transfer integral between the nearest-neighbor sites, U is the on-site Coulomb repulsion energy, $c_{i,\sigma}$ and $c^{\dagger}_{i,\sigma}$ are, respectively, the anihilation and creation operators for an electron of spin σ at the site i, $n_{i,\sigma}=c^{\dagger}_{i,\sigma}c_{i,\sigma}$ and $\langle i,j\rangle$ represents the sum over the nearest-neighbor pairs. The third term represents the coupling between the electrons and the external electrostatic field within the dipole approximation. Here, E is the magnitude of the external field, $\hat{P}=e\sum_i q_i z_i$ is the component of the polarization operator to the direction of the external field, $q_i=1-\sum_\sigma c^{\dagger}_{i,\sigma}c_{i,\sigma}$ is the charge at the site i, z_i is the component the position vector of the site i to the direction of the external field and e is the elementary electric charge. We adopt the open boundary condition.

To describe the nonlinear response to E including the strong correlation effects, we adopt the variational Monte Carlo approach [10,11]. We consider the Gutzwiller trial wave function $|\Psi\rangle=F_{\mathrm{G}}|\Phi\rangle$ [10,11], where $|\Phi\rangle$ is the ground state of the one-body part

$$H_0=-t\sum_{\langle i,j\rangle,\sigma}(c^{\dagger}_{i,\sigma}c_{j,\sigma}+c^{\dagger}_{j,\sigma}c_{i,\sigma})-E\hat{P} \tag{2}$$

and

$$F_{\mathrm{G}}=\exp\left(-\frac{1}{2}\xi\sum_i n_{i,\uparrow}n_{i,\downarrow}\right) \tag{3}$$

The Gutzwiller parameter ξ is determined so as to minimize the energy expectation value $\langle\Psi|H|\Psi\rangle/\langle\Psi|\Psi\rangle$. At least 100,000 Monte Carlo samples are used for calculating expectation values and correlation functions in all the results in this paper. We first consider the one-dimensional case. The external field is applied to the direction of the 1D chain. We show the E dependence of polarization $P=\langle\Psi|\hat{P}|\Psi\rangle/\langle\Psi|\Psi\rangle$ in Fig. 1. The system size is taken to be 64 sites. We use t,e and the lattice spacing as the units of energy, charge, and distance, respectively. VMC samples are divided into five parts, and we show the maximum and minimum values of P calculated for these parts as the error bars. We fit a curve $P=\chi^{(1)}E+\chi^{(3)}E^3$ to the VMC data using the least squares method, and the linear part $\chi^{(1)}E$ is shown by the linear lines in this figure to show the nonlinear region clearly.

As seen from this figure, $\chi^{(1)}$ weakly depends on U/t for $U/t\leq 4$, and significantly decreases as U/t is increased for $U/t\leq 4$. On the other hand, nonlinear effect is enhanced as U/t is increased. To see this more quantitatively, we show the dependence of $\chi^{(3)}$ on U/t in Fig. 2. We see that $\chi^{(3)}$ increases slowly for $U/t\leq 4$, and increases rapidly for $U\leq 6$ as a function of U/t. From exact calculation in small cluster, it is shown that there is small peak around $U/t=1$ [12], but we do not mention the peak here.

We here discuss the origin of the strong U/t dependence of $\chi^{(3)}$. For this purpose, we consider q_i induced by E, which directly determines P. We first consider the linear response region. We show the q_i at $E=0.001$ for the various values of U/t in Fig. 3.

As seen from this figure, charge density wave (CDW)-like charge density (CD) distribution is induced at $U/t=0$. This characteristic CD distribution can be understood as follows. At $U/t=0$, the ground state at $E=0$ is a metal with half-filled band, and one-electron excitations from the occupied to unoccupied levels near the Fermi energy, are induced by the weak external field. This results in the CD distribution with the wave numbers around $k=0$ and π because the Fermi wave number k_{F} is $\pm\pi/2$ in this case. This CDW arises from the instability inherent in 1D systems. The characteristic CD distribution will not be altered even when a small Coulomb gap is generated. The CD distribution weakly depends on U/t for $U/t\leq 4$. Therefore, we can conclude that the response to the weak external field can be explained within the band picture for $U/t\leq 4$. The amplitudes of q_i decrease rapidly with increasing U/t for $U/t>4$. Furthermore, the relative magnitude of the alternating component of CD with $k=\pi$ to that of the slowly varying component, decreases with increasing U/t, and the characteristic CDW-like CD distribution is not observed at $U/t=10$ as seen from Fig. 3. These results show that the

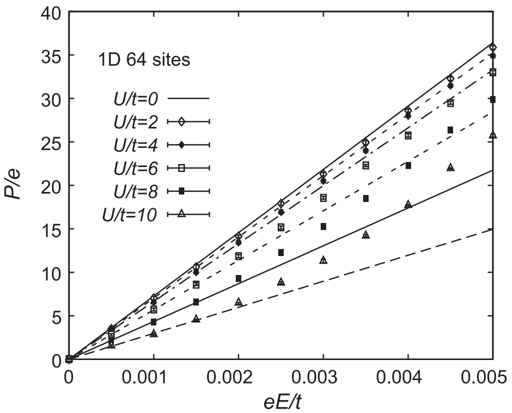

Fig. 1. The dependence of P on E for $N=64$ in the 1D case.

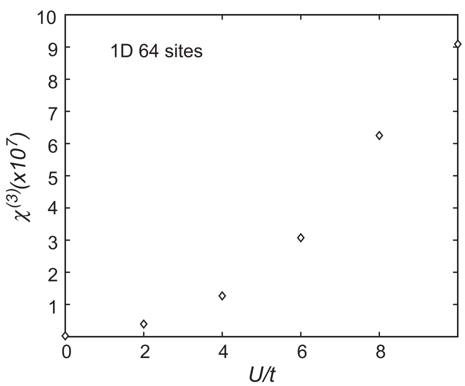

Fig. 2. The dependence of $\chi^{(3)}$ on U/t for $N=64$ in the 1D case.