Summary of thesis: Nonlinear optical responses in strongly correlated electron systems

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Nonlinear responses have been widely studied because of its great potential to reveal states in materials, such as ferroelectric domains[1], Berry curvature dipole in topological materials[2], and so on. Recently, nonlinear responses in strongly correlated electron systems are also attracting much attention, and various experiments have been conducted[3-5]. For example, in a Mott insulator Ca_2RuO_4, an unconventional gap dependence of High harmonic generation(HHG) has been observed[3], which cannot be understood by the conventional three step model. Also, in 1d ferroelectric organics $(TMTTF)_2X$($X=AsF_6$ and PF_6), photoinduced increase of short-range correlations have been observed[4], and, in a chiral magnet MnSi, nonlinear nonreciprocal transport is enhanced above the transition temperature[5]. All these experiments imply close relationship between interactions and nonlinear responses. Theoretically, one approach, which is often adopted, to nonlinear responses in strongly correlated electron systems is utilizing numerically demanding methods, e.g. exact diagonalization[6], density matrix renormalization group[7], dynamical mean field theory[8] etc., making the interpretation of results challenging. Another approach is focusing on a specific degree of freedom, e.g. excitons in semiconductors[9], where finding such an effective basis itself is a fundamentally difficult problem. Therefore, in what extent interactions and fluctuations affect nonlinear responses are not fully understood.

In Ch. 2 of this thesis, first, we discuss the gap dependence of High harmonic generation in two-level systems without interactions[10]. The simplicity of a two-level system makes it possible to investigate its gap dependence in broad parameter region varying the light-matter coupling and the rate of relaxation. As a result, we find that when the Rabi frequency is sufficiently large compared to the gap width, the HHG intensity grows as the gap width is increased, and the enhancement rate obeys a scaling law, which is independent on the incidental frequency. Also, we find that relaxation processes are not essential, but increases the visibility of the gap dependence. Furthermore, these behaviors are also confirmed in semiconducting systems. These results show that the unconventional gap dependence can appear even in non-interacting systems, and such a behavior does not originate in the peculiarity of two-level systems.

In Ch. 3 of this thesis, we discuss the dynamics of two particle correlations and its connection with nonlinear responses in the 1d interacting Rice-Mele model irradiated by an AC electric field[11]. We utilize the correlation expansion method by Fricke[12] and calculate nonlinear conductivities for the photovoltaic effect and the second harmonic generation(SHG) with a real-time approach, and propose a decomposition of conductivity into a one-particle contribution and various two-particle contributions. As a result, we find that nonlinear responses are enhanced by two-particle correlations, while the linear response is not strongly affected. Especially, the excitonic one-photon peak of the SHG conductivity is markedly enhanced by two-particle correlations. Furthermore, by the decomposition of the conductivity, we find that the "biexciton transition" correlation and its nonlinear oscillations are essential in the enhancement of the SHG. Finally, we consider the charge-charge correlations in the non-equilibrium steady state, and we find that the correlations are enhanced by driving the system with the frequency at the excitonic peak, and the intercell correlation can even exceed the intracell one by the irradiation of the external field. These results imply the importance of two-particle correlations in nonlinear responses and the possibility to control fluctuations of correlated systems by the external field. Also, our approach can be easily extended to other systems, and can be utilized to clarify what kind of interactions and correlations are essential to describe nonlinear optical properties of a system.

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