Theoretical study on mechanism of solid-state high-harmonic generation

論文の内容の要旨

論文題目

Theoretical study on mechanism of solid-state high-harmonic generation
(固体高次高調波発生のメカニズムに関する理論的研究)

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The advent of the laser in 1960 has opened a new era of nonlinear optics in the field of optical science. In the nonlinear regime, the optical properties of materials are no longer independent of the field strength. Today, advances in ultrafast intense laser techniques have enabled us to enter a new stage beyond nonlinear optics: strong-field physics, where perturbative treatments of the field are no longer applicable. In particular, high-harmonic generation (HHG) from gas-phase atoms and molecules has been one of the main targets of research for three decades, which has led to successful applications such as attosecond pulse and coherent keV x-ray generations as well as powerful means to observe and manipulate ultrafast electron dynamics. Many features of gas-phase HHG can be intuitively and even quantitatively explained by a semiclassical three-step model.

Recently, solid-state materials have emerged as a new playground for strong-field physics. In particular, since its first discovery in 2011, many experimental observations of high-harmonic generation from solid-state materials, or *solid-state high-harmonic generation*, have been reported. The extension of the concepts and methods that have been developed so far in the gas-phase HHG is expected to offer potential scientific and technological opportunities in solids. The application of spectroscopic techniques in gas-phase HHG to solid-state systems may provide new means to probe the electronic structures of insulators and semiconductors. Understanding the electron excitation process under intense electric fields would be critical to future applications in ultrafast optical current control and be important as the initial process of laser material processing.

As this field is in its infancy, the radiation mechanism is still under intensive discussions. Experimentally, several features unique to solids have been discovered such as the linear cutoff-energy scaling with field strength and a sudden transition from a single- to a multiple-plateau spectral structure. These experimental results have required the theoretical development of new methods and models that incorporate intrinsic characters of solids.

Along with experiments, numerical approaches have played an important role in strong-field science to reveal underlying processes. Especially in solids, which have a plethora of effects that may overshadow pure electron responses inside materials, numerical methods are indispensable for revealing essential processes underlying the phenomena. Therefore, the development of numerical approaches to describe the many-electron system in solids in the strong-field regime has been in high demand.

In this dissertation, I theoretically study the mechanism of the solid-state HHG by developing numerical methods, focusing on the following topics:

(i) Independent-electron dynamics in HHG in solids

The experimentally observed unique features of the solid-state HHG require the development of new methods and models that incorporate the intrinsic properties of solids. Although pioneering studies have discussed several models, they have considered only a two-

band model with a single valence band (VB) and a single conduction band (CB). More recently, the importance of considering multiple energy bands has been pointed out by several authors.

In this work, we study HHG from solids using a one-dimensional model crystal by numerically solving the time-dependent Schrödinger equation (TDSE) within the independent-electron approximation. By solving the TDSE directly on a spatial grid, which involves multiple-band contribution in a natural manner, we have successfully reproduced harmonic spectra with several unique features such as the linear scaling of cutoff energy and a sudden transition from a single- to a multiple-plateau structure [Fig. 1(a)]. With increasing field strength $\mathcal{E}_0$, the second and third plateaus suddenly appear at $\mathcal{E}_0 \approx 1.4$ V/nm, and moreover, another cutoff jump is seen at $\mathcal{E}_0 \approx 2.8$ V/nm. In addition, defining A_{peak} as the maximum peak-to-valley amplitude of the vector potential $A(t)$ [Fig. 1(c)], we have found that these jumps well coincide with $A_{\text{peak}} = \pi/a = 0.393$ (atomic unit) and $2\pi/a = 0.786$ (atomic unit), with a being the lattice constant.

Based on the simulation results, we have proposed and discussed a simple model that consists of three types of electron dynamics: (i) tunneling excitation to an adjacent upper band at the minimum band gap (MBG); (ii) intraband displacement expressed by $k(t) = k_0 + A(t)$ with k_0 the initial crystal momentum; (iii) photon emission upon interband transition to the initial VB, whose energy is equal to the particle-hole energy $\Delta\varepsilon_{mn}\big(k(t)\big) = \varepsilon_{mk(t)} - \varepsilon_{nk(t)}$ between the band m and n, corresponding to before and after the time evolution respectively. Two typical trajectories are depicted in Fig. 1(b). An electron initially in the VB undergoes intraband displacement and is excited at the MBG to the first CB, denoted as $t = t_0$. The subsequent momentum displacement in the first CB is given by $A(t) - A(t_0)$, where $|A(t) - A(t_0)|$ is bounded by A_{peak}. Hence, if $A_{\text{peak}} < \pi/a$, no excited electrons can reach the next MBG. Electrons oscillate in the first CB, which forms a single plateau in the high-harmonic spectra. On the other hand, if $A_{\text{peak}} > \pi/a$, some electrons can reach the next MBG, be promoted to the second CB, and further climb up to higher and higher CBs by repeating the intraband displacement and interband tunneling, leading to the formation of multiple plateaus. The cutoff positions as well as the time-frequency structure of HHG [Figs. 1(d) and (e)] can be deduced by tracing all the trajectories starting from different initial crystal momenta. Based on the stepwise excitation process in the momentum space, we predict yet another difference from gas-phase HHG: that the position of the highest cutoff energy depends on not only the wavelength and field strength of the pulse but also its duration.

Our trajectory analysis can be viewed as a solid-state and momentum-space counterpart with multi-band extension of the three-step model in the gas-phase HHG. It provides a unified basis for understanding HHG in gas-phase and solid-state materials. This offers a clear physical insight into the coherent electron dynamics of its independent nature and serves as a benchmark to identify the effects of electron correlation, relaxation, dephasing, impurity, distortion, etc., in real experiments.

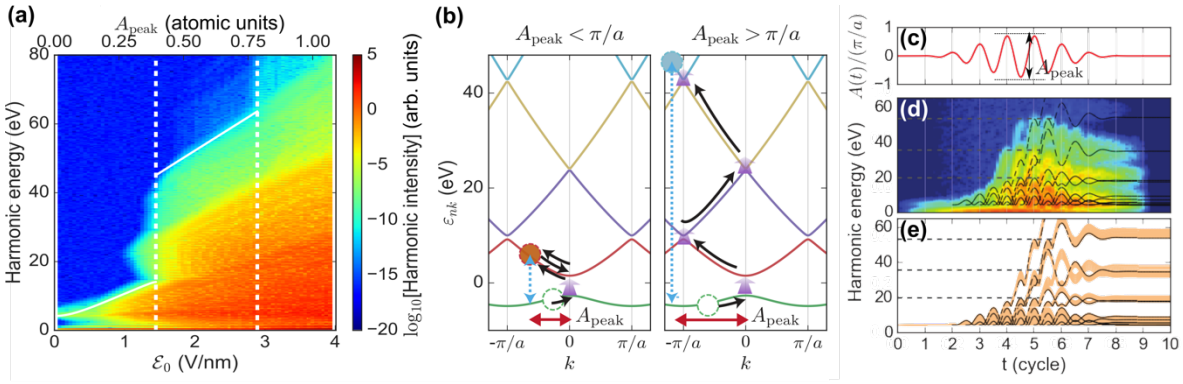

Figure 1: (a) Simulated HHG spectra under three-cycle pulse field as functions of field strength $\mathcal{E}_0$ and the maximum peak-to-valley amplitude A_{peak} of the vector potential $A(t)$. The vertical dashed white lines represent $A_{\text{peak}} = \pi/a$ and $2\pi/a$ and white oblique lines the cutoff energy positions predicted by our model. (b) Pictorial representation of

our model for the electron dynamics in the momentum space when (left) $A_{\text{peak}} < \pi/a$ and (right) $A_{\text{peak}} > \pi/a$. (c-e) The temporal evolution of harmonic spectrum for $A_{\text{peak}} = 0.57 > \pi/a$. (c) The waveform of the vector potential $A(t)$ normalized to π/a. (d) Time-frequency analysis of HHG. (e) Electron energy trajectories based on the model. The equivalent trajectories and characteristic energy positions are shown by black and gray dashed lines in (d) and (e) to help the comparison.

(ii) Multielectron effects on HHG in solids

Unlike the ionized electrons from isolated atoms and molecules, electrons (tunnel-)excited to a CB move through the background of other electrons. While most of the previous discussions are based on the independent-electron approximations, the role of multielectron effects in solids in strong-field regime is largely unexplored.

In this work, we study the effects of the electron-hole interaction (EHI) on the solid-state HHG by developing the time-dependent Hartree-Fock (TDHF) simulation methods. By numerically solving the TDHF equations for a one-dimensional model crystal, we have found a qualitative change in the harmonic spectra from what is obtained with the independent-electron simulations [Fig. 2(a) and (b)]. In particular, the second plateau appears at $A_{\text{peak}} \approx 0.5 \ll \pi/a = 0.87$, a much smaller field strength than for the independent-electron case, which marks a striking manifestation of EHI.

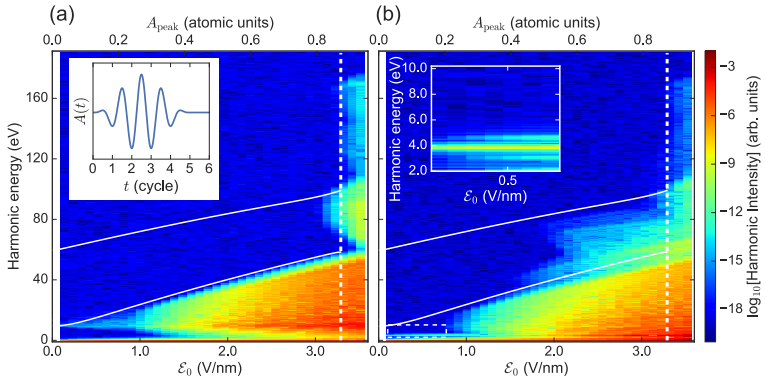

Figure 2: Harmonic spectra as functions of the field amplitude $\mathcal{E}_0$ and corresponding A_{peak} from (a) independent-electron and (b) TDHF simulations. The dashed vertical lines denote $A_{\text{peak}} = \pi/a = 0.87$. The solid lines are the cutoff energy positions predicted from the trajectory analysis. The inset in (a) shows the waveform of $A(t)$ used in the simulations.

In order to reveal the origin of this qualitative change, we consider dynamics of the time-dependent electron orbital $\psi_{bk_0}(x,t)$ whose initial state is the self-consistent eigenstate $\phi_{bk_0}(x)$ of field-free Hartree-Fock Hamiltonian with eigenenergy ε_{bk_0}. We expand $\psi_{bk_0}(x,t)$ with the accelerated Bloch states, or the Houston states, $\tilde{\phi}_{nk_0}(x,t) = e^{-iA(t)x}\phi_{nk(t)}(x)$ of initial VB ($n = v$) and the first CB ($n = c$) with $k(t) = k_0 + A(t)$,

$$\psi_{bk_0}(x,t) \approx a^v_{k_0}(t)e^{-i\int \varepsilon_{vk(t')}dt'}\tilde{\phi}_{vk_0}(x,t) + a^c_{k_0}(t)e^{-i\int \varepsilon_{ck(t')}dt'}\tilde{\phi}_{ck_0}(x,t),$$

We assume that population transfer from the VB to CBs are small, that is, $a^v_{k_0}(t) \approx 1$ and $a^c_{k_0}(t) \ll 1$. Employing some approximations used in the semiconductor Bloch equations, which has been widely used to describe optical responses of semiconductors, the equation of motion for the complex amplitude $a^c_{k_0}(t)$ is given by

$$i\frac{d}{dt}a^c_{k_0}(t) \approx e^{i\int \Delta\varepsilon_{cv}\left(k(t')\right)dt'}\left[\mathcal{E}(t)\xi^{cv}_{k(t)} - \sum_{q\neq 0} v_{-q}D^{cv}_{k(t)+q}(t)\right],$$

where v_q is the spatial Fourier transform of the interelectronic Coulomb potential, $\xi^{mn}_k = i\langle u^0_{mk}|\nabla_k u^0_{nk}\rangle$ with $u^0_{nk}(x)$ being the lattice periodic part of the initial Bloch state $\phi_{nk}(x)$, and

$D_k^{cv}(t) = a_{k_0}^c(t)e^{-i\int \Delta\varepsilon_{cv}\left(k(t')\right)dt'}$ is the electron-hole polarization at a crystal momentum $k(t)$. The first term describes the independent-electron dynamics, depicted by the momentum-space trajectory analysis [Fig. 3(a)]. The second term stems from the electron-hole interaction in the TDHF Hamiltonian, which indicates that the electron-hole polarization created at a remote crystal momentum $k(t) + q$ can induce quasi-resonant excitation because it has much higher frequency $\Delta\varepsilon_{cv}(k(t) + q)$ than the electric field $\mathcal{E}(t)$. This means that the Coulomb interaction from the electron-hole polarization at the minimum band gap mediates excitation of distant valence-band electrons, which we call hauling-up effects [Fig. 3(b)]. In other words, this hauling-up process offers a shortcut route for VB electrons far from the minimum band gap to climb up to the first CB assisted by an electron-hole polarization formed at a different k-point. This mechanism is supported by the time-frequency structure of HHG and final band populations.

If we shift our eyes back to the gas-phase HHG, the influence of the Coulomb potential from the parent ion, neglected in the strong-field approximation, may somewhat correspond to that of EHI. However, it hardly affects qualitative features of harmonic spectra. Our results suggest that the solid-state HHG mechanism involves much more complicated process than the gas-phase, and hence, may offer possible opportunities to reveal correlation in ultrafast electron dynamics in solids. The present study will pave the way toward the ultimate goal of revealing correlations in ultrafast electron dynamics in solids.

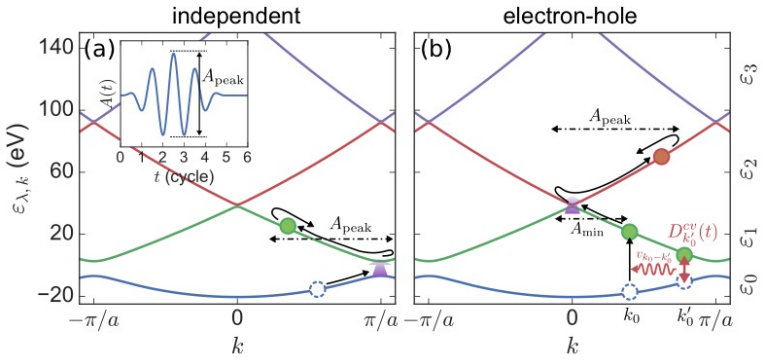

Figure 3: Pictorial representation of electron dynamics in the momentum space (a) within the independent-electron trajectory analysis and (b) involving the hauling-up effect arising from interelectronic interaction in the TDHF Hamiltonian.