Theoretical study on the laser-matter nonlinear interaction

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Recently, a high-order harmonic generation (HHG) in the solid state has attracted great interests as a new short wavelength light source and an observation method of the physical properties [1]. Although HHG in a solid is of interest in electronic excitations, details of the physical process are unclear. On the other hand, due to the development of attosecond science, transient absorption spectroscopy is expected to clarify the nonlinear ultrafast phenomena [1]. However, a new physical interpretation is important as prior models have not reproduced the experimental results.

New theories and numerical approach are needed to elucidate these nonlinear and ultrafast electronic dynamics. The description of the electronic ground state can be calculated by density functional theory (DFT). On the other hand, the electronic dynamics can be described by time-dependent density functional theory (TDDFT) [3], which has successfully reproduced many nonlinear ultrafast phenomena. In this study, we present numerical calculations for HHG in solid and transient spectroscopy by an attosecond light pulse.

Figure 1 shows the numerical result for HHG in α-quartz when the laser wavelength is set to 800 nm. The laser intensity in the α-quartz is assumed to be $8\text{x}10^{13}$ W/cm^2. We set the polarization direction parallel to the optical axis. The applied electric field (red-dashed line) and the induced electronic current (blue line) are shown in Fig. 1(a), while Fig. 1(b) presents the Fourier transformation of the current corresponding to HHG.

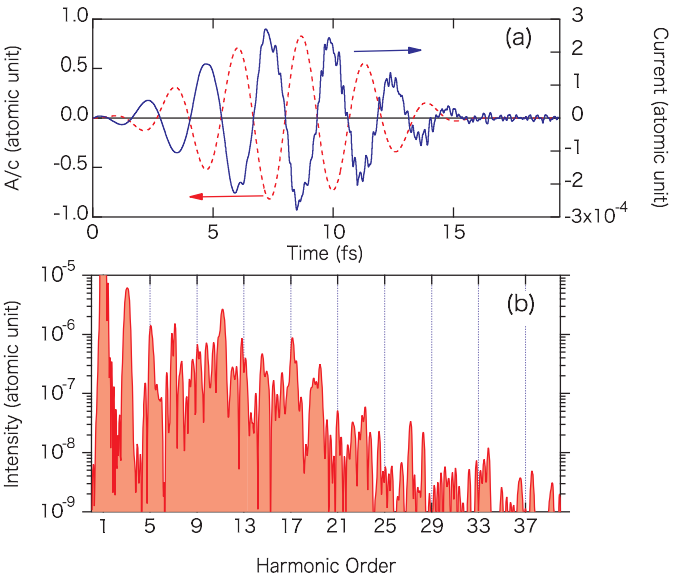

Figure 1: (a) Laser field and the induced current. (b) Spectrum of the electronic current.

To understand HHG in solids, it is important to elucidate the contribution of the inter- and intra-band transitions. Figure 2 shows HHG by the full calculation (red line) and the intra-band transition (blue-filled). The contribution of the intra-band transition is clearly a minor effect. From the comparison between HHG spectrum and the band structure, HHG is attributed to the recombination between electrons around the bottom of the conduction band and holes. The above results are reported in Physical Review B [4].

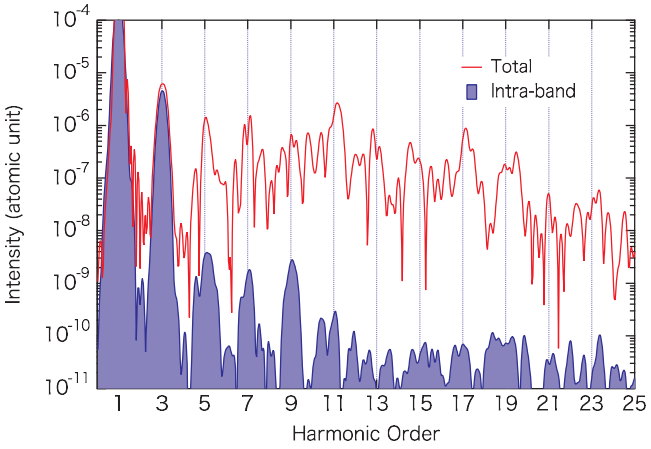

Figure 2: (a) Laser field and the induced current. (b) Spectrum of the electronic current.

We studied numerical pump-probe transient spectroscopy using TDDFT and analytical theory. Our results showed an intense subcycle modulation of the optical properties under an intense linear polarized laser [5]. In this study, we extend the theory to elliptic polarized lasers [6].

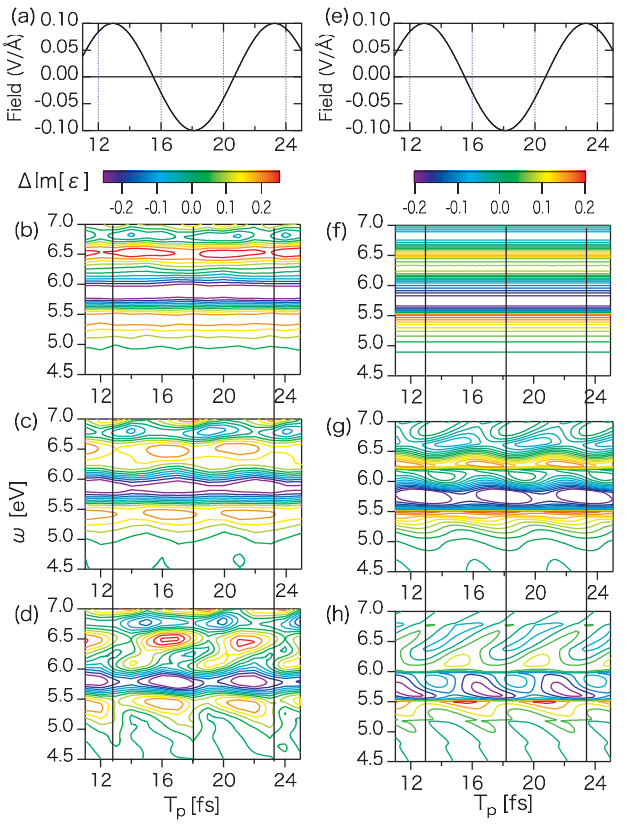

Figure 3: Polarization dependence in transient spectroscopy. (a) and (e): Electric field of the linearly polarized pump. (b) and (f): Circular polarization. (c),(g) Elliptic polarization. (d) and (h): Linear polarization

Figure 3 shows the results by TDDFT (left panels) and analytical theory (right panels) for diamond. Panels (a) and (e) show the linear polarized pump laser field, where the vertical solid lines represent the peak of the electric field. The modulation of the imaginary part of the dielectric function is shown as the contour plot in panels (b)-(d) and (f)-(h). In our calculations, the optical band gap is 5.6 eV. The intense subcycle modulation under a linear polarized laser (panels (d) and (h)) becomes weaker as the ellipticity increases. Of particular note is that the time-dependence disappears in the circularly polarization case ((b) and (f)). The analytical theory shows a good qualitative agreement with TDDFT.

The derived analytic formula corresponds to the response of the Floquet states. Therefore, the subcycle transient spectroscopy can be understood as the response of the many Floquet states (Dressed states), including their phase at the probe time. This effect has been observed experimentally by attosecond spectroscopy [7]. A similar effect has been found in transient spectroscopy of the excitonic state under a terahertz field [8].

We elucidated the physical processes in HHG and transient spectroscopy by numerical simulations. Some experiments indicated a laser induced subcycle modulation. We would like to extend the program to explore new functional materials. Our numerical method is also suitable to study the initial state of laser processing. We are planning to explore the development of a new efficient processing method.

Acknowledgement

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