Ab-initio simulation of electron energy distribution in an insulator excited by an intense laser pulse

Yasushi SHINOHARA[1,2]

[1]*Photon Science Center,*

The University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113-8656.

[2]*NTT Basic Research Laboratories,*

NTT Corporation, 3-1 Morinosato Wakamiya, Atsugi, Kanagawa 243-0198.

We have investigated responses of insulators exposed to strong light fields via time-dependent density-functional theory (TDDFT) [1]. When the field strength of light is comparable to the material's internal strength, the material exhibits extremely nonlinear responses that cannot be described by perturbative series. The only viable option to describe these responses is a direct solution of the time-dependent Kohn-Sham equation in real-time. The SALMON-TDDFT code [2] provides opportunities for solving the Kohn-Sham equation for crystalline solids, molecules, and atoms. We used system B at SCC-ISSP.

To simulate subsequent phenomena after the strong field excitation, we need to consider multi-physical nature of spatiotemporal evolution beyond electron quantum degree of freedom such as thermalization over whole subsystems, energy/momentum flows in microscopic spatial range, phase transition and so on. We propose an electron energy distribution after excitation to bridge from TDDFT to a more macroscopic theoretical framework. The electron distribution is obtained by projecting the time-dependent orbitals $\psi_{c\boldsymbol{k}}(t)$ onto the ground state orbitals $\phi_{b\boldsymbol{k}}$:

$$f_{b\boldsymbol{k}}(t) = \sum_{c} |\langle \phi_{b\boldsymbol{k}} | \psi_{c\boldsymbol{k}}(t) \rangle|^2.$$

The occupation $f_{b\boldsymbol{k}}$ at crystal momentum $\boldsymbol{k}$ is interpreted as particle (hole) distribution for b belongs to a conduction (valence) band. We obtain laser-excited (LE) particle/hole distributions as

$$f_p^{\mathrm{LE}}(E) = \sum_{b(\in \mathrm{val.})\boldsymbol{k}} f_{b\boldsymbol{k}}(t = t_{\mathrm{end}})\, \delta(E - \epsilon_{b\boldsymbol{k}}),$$

$$f_h^{\mathrm{LE}}(E) = \sum_{b(\in \mathrm{cond.})\boldsymbol{k}} f_{b\boldsymbol{k}}(t = t_{\mathrm{end}})\, \delta(E - \epsilon_{b\boldsymbol{k}}),$$

where $\epsilon_{b\boldsymbol{k}}$ is the eigenvalue of the initial Kohn-Sham Hamiltonian. We obtain particle and hole effective temperatures, T_p, T_h, as characteristics of the distribution by fitting such that the particle (hole) distribution gives minimum error to a Fermi-Dirac distribution $f(E, \mu, T)$ at the effective temperature:

$$\arg\min_{T} \left(\int dE \left| f_{p/h}^{\mathrm{LE}}(E) - f\left(E, \mu_{p/h}, T\right) \right| \right) = T_{p/h},$$

where chemical potential is determined by a fact that energy integrals of the distribution give the same value, namely number of particle/hole conservation.

We perform TDDFT simulation for α-quartz with a driving field that have 1.60 eV, 13 fs pulse duration. The temperature depending on peak intensity are presented in Fig. 1. The Keldysh parameter, a characteristic value whether tunneling picture is valid or not, is equal to unity when the intensity is equal to 10 TW/cm^2. The temperatures monotonically increase as a function of the peak intensity. The sum of temperatures, interpreted as the averaged kinetic energy of the particle-hole pair, is 17.8 eV at 100 TW/cm^2. The value is close to the ponderomotive energy 19 eV evaluated with the reduced effective mass of 0.3. This coincidence invokes that kinetic energy gain is understood as free particle-hole motion after the tunneling ionization.

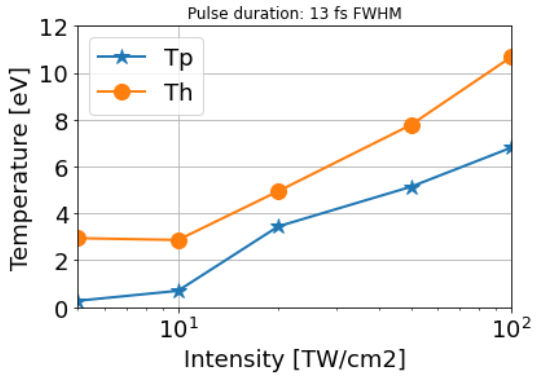

Fig. 1: Evolution of particle- and hole-temperatures.

References

[1] Erich Runge and E. K. U. Gross, Phys. Rev. Lett. **52**, 997 (1984).

[2] https://salmon-tddft.jp/, M. Noda, S.A. Sato, et al., Computer Physics Communications. **235**, 356 (2019).