Time-dependent density functional theory for ultrafast electron dynamics at solid surfaces

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Abstract: We present first-principles simulation based on the time-dependent density functional theory for electron dynamics in crystalline Si under irradiation of intense and ultrashort laser pulse. Including pump and probe pulses in the applied electric field, we numerically investigate dielectric properties of bulk Si with dense electron-hole excitations caused by the intense pump pulse. We find the contribution of the excited carrier to the dielectric function is reasonably described by the Drude model at least for the real part.

1. Introduction

Irradiation of intense and ultrashort laser pulse on solid surface induces strong excitations of electrons in femtosecond time scale. To understand properties of excited solid surface, ab-initio numerical simulation will be useful. We have developed a theoretical framework for electron dynamics in a unit cell of crystalline solid based on the time-dependent density functional theory (TDDFT). We solve the time-dependent Kohn-Sham (TDKS) equation in real-time and real-space [1-4].

In this presentation, we report numerical pump-probe calculation for bulk Si to study dielectric properties of bulk Si excited by the intense pump pulse. Making Fourier analysis for the probe pulse, we can extract dielectric function of excited Si.

2. Frameworks

We investigate electron dynamics in a crystalline solid under a spatially uniform, time-dependent electric field by solving the following TDKS equation:

$$i\hbar\frac{\partial}{\partial t}\psi_i(\vec{r},t) = \hat{h}_{KS}(t)\psi_i(\vec{r},t),$$

$$\hat{h}_{KS}(t) = \frac{1}{2m}\left(\vec{p}+\frac{e}{c}\vec{A}_{ext}(t)\right)^2 + \hat{V}_{ion} + e^2\int d\vec{r}\,'\frac{n(\vec{r},t)}{|\vec{r}-\vec{r}\,'|} + V_{XC}[n(\vec{r},t)],$$

where $\psi_i(\vec{r},t)$ are Bloch orbitals, $\vec{A}_{ext}(t)$ is the vector potential describing the applied electric field, $\hat{V}_{ion}$ is the electron-ion interaction potential, $n(\vec{r},t)$ is the electron density, and $V_{XC}[n(\vec{r},t)]$ is the exchange-correlation potential. We evolve the orbitals starting from the ground state solution.

In order to investigate dielectric properties of crystalline solid excited by a laser pulse, we perform *numerical pump-probe experiments* in which electric fields of pump and probe pulses are included in $\vec{A}_{ext}(t)$. The intense pump pulse induces dense electron-hole excitations and the probe pulse brings information on the dielectric properties of the excited Si.

The dielectric function of excited Si can be extracted from the response of the probe pulse in the following way. We take Fourier transforms of the applied probe pulse, $E_{probe}(t)$, and the induced electron current, $J_{probe}(t)$. We then obtain conductivity $\sigma(\omega)$ and dielectric function $\epsilon(\omega)$ by

$$J_{probe}(\omega) = \sigma(\omega)E_{probe}(\omega)$$

$$\epsilon(\omega) = 1 + \frac{4\pi i}{\omega}\sigma(\omega),$$

We repeat this analysis changing parameters of pump and probe pulses such as intensity and frequency to obtain dielectric properties of excited Si.

3. Results & Conclusion

In Fig. 1, we show typical time profiles of the pump-probe electric fields and the electron current induced by the pulses. The peak intensity of the pump pulse is set to $2\times10^{12}\mathrm{W/cm^2}$, mean frequency to $\hbar\omega = 1.55$ eV, and the pulse length to about 10 fs. We find oscillatory current which is almost proportional to the applied laser pulses. One notes that the current does not vanish even after the pump pulse ends. The oscillatory current during the applied laser pulse originates from a virtual electronic excitation, while the current after the laser pulse ends indicates real electronic excitation caused by multiphoton absorption process. The number of excited electrons is evaluated to be 0.015 electrons per Si atom in this case.

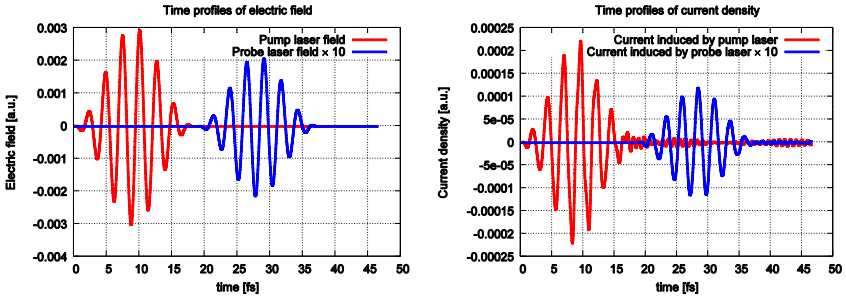

Figure 1, Left panel shows the time profiles of pump and probe electric fields applied to bulk Si. Right panel shows the electron current induced by the pump and probe pulses.

In Fig. 2, we show the dielectric function of the crystalline Si excited by the pump pulse, the real-part in the left panel and the imaginary part in the right-panel. The numerical setting of the pump pulse is the same as that of Fig. 1. From the left panel, we find the dielectric function shows metallic behavior at low frequency, $\varepsilon(\omega) \propto -1/\omega^2$. We fit the calculated dielectric function as a sum of that of the ground state and that of the excited electrons which we assumes Drude model. The result is included in Fig. 2. In the Drude model, there are three parameters: electron-hole density n_{e-h}, electron-hole reduced mass m^*, and damping time τ. We use calculated value for n_{e-h} and treat other two parameters as fitting parameters. From Fig. 2, one can see that the calculated dielectric function is well fitted by the Drude model, at least for the real-part. We thus conclude that dielectric properties of excited Si may be reasonably described by the sum of the dielectric functions of the Si in the ground state and that of the excited carrier electrons in the Drude model.

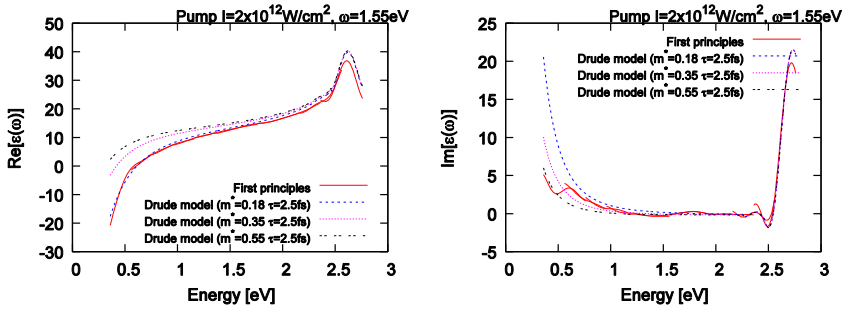

Figure 2, Dielectric function of the crystalline Si in the numerical pump-probe experiment. Fits by Drude model are also shown.

4. References

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