Semi-classical approach for laser-metal interaction

OTOBE Tomohito

Ultrafast Dynamics Group, Department of Advanced Photon Research

The comprehensive modeling of laser material machining [1-3] is complex, with multi-scale time and space, multi-phase, and possibly chemical reactions. For laser processing of metals, a two-temperature model (TTM) has been proposed as the simplest continuum model [4]. In the TTM, empirical parameters are employed for all material properties, and the penetration depth models the dynamics of the electromagnetic field. The most critical assumption of the TTM is the quasi-equilibrium of the electron temperature and the electron-lattice interaction term.

Recently, the first-principles approach employing time-dependent density functional theory (TDDFT) [5] has been applied to laser-matter interactions [6,7]. Although TDDFT offers a compromise between accuracy and computational feasibility, its computational cost remains high. Electron-electron collisions (which are not included in TDDFT) play an important role in the laser-metal interactions.

The Vlasov equation has been employed in nuclear physics and electron dynamics in metal clusters to describe the collision process of fermi particles [8-10]. Because the Vlasov equation treats the distribution function of space, momentum, and time, it requires tremendous computational resources. In general, the computational cost is reduced by assuming that the distribution function is the summary quasi-particles.

The time-dependent Kohn–Sham equation [5] is the fundamental equation of the TDDFT.

$$i\hbar\frac{\partial\phi_i}{\partial t} = H_{KS}\phi_i \qquad 1$$

Here:

$$H_{KS}[\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t})] = -\frac{\hbar^2}{2\mathrm{m}}\nabla^2 + \mathrm{V_{eff}}[\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t})] \qquad 2$$

where H_{KS} was the Kohn–Sham Hamiltonian, m the electron mass, $\mathrm{V_{eff}}$ the effective potential, and $\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t})$ the time-dependent electron density.

$$\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t}) = \sum_{i=1}^{N}|\phi_i(\vec{r},t)|^2 \qquad 3$$

Further, the von Neumann equation governed the time-evolution of the density matrix $\hat{\rho}$

$$\frac{\partial\hat{\rho}(\vec{r},\vec{r}',t)}{\partial t} = -\frac{i}{\hbar}\left[\hat{H}_{KS},\hat{\rho}(\vec{r},\vec{r}',t)\right]. \qquad 4$$

By performing the Wigner transformation and taking the limit $\hbar\to 0$, the density operator was mapped onto a real function $f(\vec{r},\vec{p},t)$, which obeyed the Vlasov equation:

$$\frac{\partial}{\partial t}f(\vec{r},\vec{p},t) = -\frac{\vec{p}}{m}\cdot\nabla_{\vec{r}}f(\vec{r},\vec{p},t) + \nabla_{\vec{r}}V_{eff}\cdot\nabla_{\vec{p}}f(\vec{r},\vec{p},t), \qquad 5$$

Here, $f(\vec{r},\vec{p},t)$ was interpreted as the electron distribution function in phase space.

The effective potential was a function of the electron density $\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t})$ and decomposed into

$$\mathrm{V_{eff}}[\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t})] = V_{Cou}[\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t})] + V_{xc}[\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t})] + V_{ext}(\vec{r},t), \qquad 6$$

with the exchange-correlation potential V_{xc} [11], external field potential V_{ext}, and

$$V_{Cou}[\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t})] = \sum_j V_{ps}(\vec{r}-\vec{r}'_j) + V_H[\mathrm{n_e}(\vec{\mathrm{r}},\mathrm{t})], \qquad 7$$

where V_{ps}, V_H, and j denoted the ionic pseudopotential, Hartree potential, and label of ions, respectively.

We employed a modified Heine–Abarenkov-type local pseudopotential for V_{ps} with parameters to reproduce the all-electron density functional theory calculations [12]. V_H was evaluated by solving the Poisson equation. V_{xc} was the exchange-correlation potential in the local density approximation. The laser-electron interaction was described in the length gauge.

We approximated the six-dimensional time-dependent function with the summation of the pseudo particle (PP):

$$f(\vec{r},\vec{p},t) = \frac{1}{N_s}\sum_{k=1}^{N_{pp}} g_r(\vec{r}-\vec{r}_k(t))g_p(\vec{p}-\vec{p}_k(t)) \qquad 8$$

Here, k was the label of the PP, and N_{pp} the total number of PPs defined as $N_{pp} = N_sN_e$, where N_s and N_e were the number of PPs per electron and the total number of electrons, respectively.

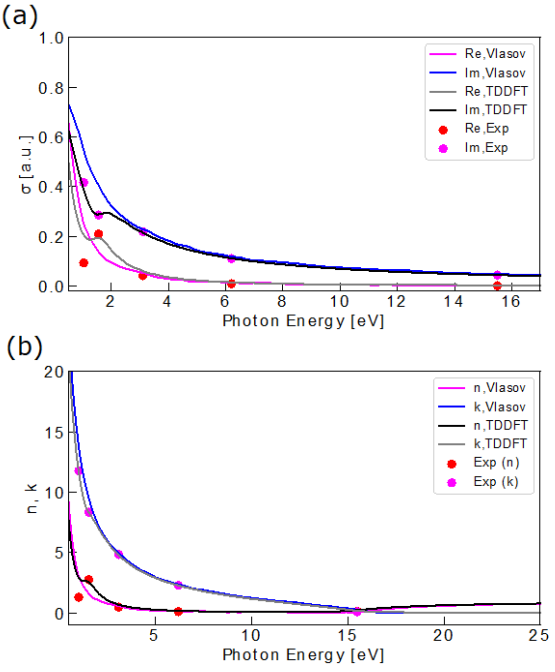

Fig. 1 (a) Conductivity and (b) refractive indexes of bulk-Al with the Vlasov equation and TDDFT [15]. The filled circles indicate the experimental results [13].

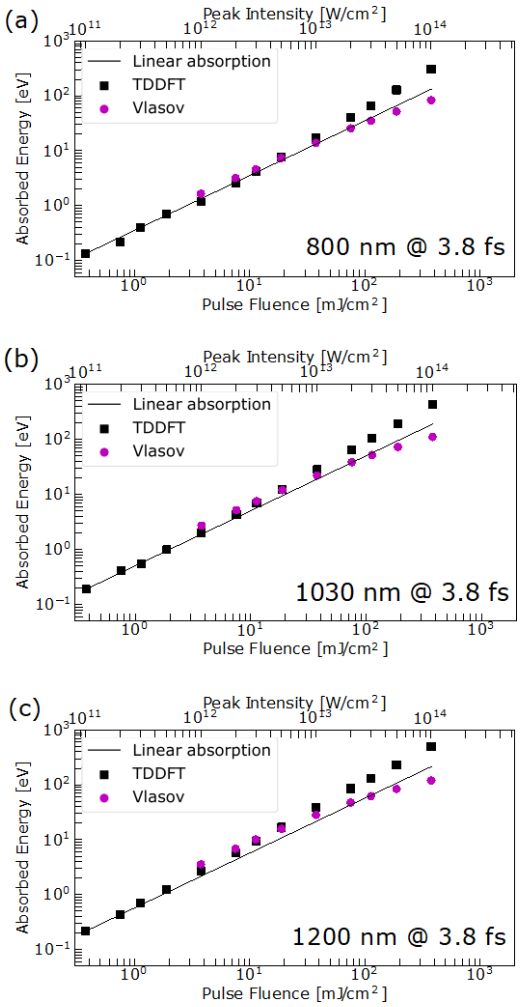

Fig. 2 Laser fluence dependence of the energy absorption with a wavelength of (a) 800 nm, (b) 1,030 nm, and (c) 1,200 nm.[15]

We defined the smoothing functions for position $g_r(\vec{r})$ and momentum $g_p(\vec{p})$ in the Gaussian form. The Newton equation described the motion of the PP under an effective potential with a periodic boundary condition as follows:

$$\frac{d\vec{r}_k}{dt} = \frac{\vec{p}_k}{m} \qquad 9$$

$$\frac{d\vec{p}_k}{dt} = -\int d\vec{r}\, V_{eff}(\vec{r}) \nabla_{\vec{r}} g_r(\vec{r}_k(t) - \vec{r}) \qquad 10$$

The dynamics of PPs were performed by the Verlet method [14].

We discussed the complex optical conductivity, refractive index, extinction coefficient, and reflectivity as functions of photon energy [15]. Despite the simplicity of the Vlasov approach, the results agreed with the TDDFT results and experimental values (Fig. 1). The peak and dip at approximately 1.5 eV in the TDDFT results were due to inter-band transition, which was not reproduced by the present Vlasov approach because the latter considered only the free-electron dispersion.

Fig. 2 shows the absorbed energy as a function of laser intensity with laser wavelengths of (a) 800 nm, (b) 1,030 nm, and (c) 1,200 nm [15]. The triangles indicated the results obtained using TDDFT, and the circles indicated the results obtained using the Vlasov equation. The scaled one-photon absorption was indicated by the solid line for the guiding eye. At all wavelengths, the Vlasov equation agreed with TDDFT below the intensity of 1×10^{13} W/cm^2. Above 1×10^{13} W/cm^2, Vlasov underestimated compared with TDDFT.

Here, we proposed a semi-classical approach for the laser-metal interactions employing the Vlasov equation. The master equation and potential were based on the Kohn–Sham Hamiltonian with a local density approximation. We employed PP to reproduce the continuous density distribution function in the real and momentum spaces. Our results agreed with TDDFT for the weak and intense laser field regimes. In particular, the Vlasov equation reproduced the optical properties of aluminum without artificial parameters. The advantages of the Vlasov equation was its low computational cost (approximately 1/50 of TDDFT) and high scalability. Assuming Pauli blocking, we included the electron-electron collision process in the materials. Further, the collision between PP and the pseudo-potential ensured semiclassical molecular dynamics.

Acknowledgments

This research was supported by the MEXT Quantum Leap Flagship Program (MEXT Q-LEAP) (Grant No. JPMXS0118067246. The numerical calculations were performed on the supercomputers Oakbridge-CX, Sekirei, oh take (University of Tokyo), and SGI ICE X at the Japan Atomic Energy Agency (JAEA). We would like to thank Editage (www.editage.com) for English language editing.

References

1. C. Schäfer, H. M. Urbassek, and L. V. Zhigilei, Phys. Rev. B **66**, 115404 (2002)
2. C. Kerse, H. Kalaycıoglu, P. Elahi, B. Çetin, D. K. Kesim, Ö. Akçaalan, S. Yavaş, M. D. Aşık, B. Öktem, H. Hoogland, R. Holzwarth, and F. Ö. Ilday, Nature (London) **537**, 84 (2016)
3. R. R. Gattass and E. Mazur, Nat. Photonics **2**, 219 (2008)
4. S. I. Anisimov, B. L. Kapeliovich, and T. L. Perel'man, JETP **39**, 375 (1974)
5. E. Runge and E. K. U. Gross, Phys. Rev. Lett. **52**, 997 (1984)
6. T. Otobe, M. Yamagiwa, J. -I. Iwata, K. Yabana, T. Nakatsukasa, and G. F. Bertsch, Phys. Rev. B **77**, 165104 (2008)
7. M. Noda, S. A. Sato, Y. Hirokawa, M. Uemoto, T. Taleuchi, S. Yamada, A. Yamada, Y. Shinohara, M. Yamaguchi, K. Iida, I. Floss, T. Otobe, K.-M. Lee, K. Ishimura, T. Noku, G. F. Bertsch, K. Nobusada, and K. Yabana, Comp., Phys. Commun. **235**, 356 (2019)
8. A. Domps, P. -G. Reinhard, and E. Suraud, Ann. Phys. **260**, 171 (1997)
9. L. Plagne, J. Daligault, K. Yabana, T. Tazawa, Y. Abe, and C. Guet, Phys. Rev. A **61**, 033201 (2000)
10. T. Fennel, G. Bertsch, and K.-H. Mwiwes-Broer, Eur. Phys. J. D **29**, 367 (2004)
11. J. P. Perdew and A. Zunger, Phys. Rev. B **23**, 5048 (1981)
12. C. Fiolhais, J. P. Perdew, S. Q. Armster, J. M. MacLaren, and M. Brajczewska, Phys. Rev. B **51**, 14001 (1995)
13. A. D. Rakić, Appl. Opt. **34**, 4755 (1995)
14. L. Verlet, Phys. Rev. **159**, 98 (1967)
15. M. Tani, T. Otobe, Y. Shinohara, and K. L. Ishikawa, Phys. Rev. B **104**, 075157 (2021)