

Maxwell-Vlasov-Uehling-Uhlenbeck simulation for coupled laser-electron dynamics in a metal irradiated by ultrashort intense laser pulses

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The description of electron-electron scattering presents challenges in the microscopic modeling of the interaction of ultrashort intense laser pulses with solids. We extend the semiclassical approach based on the Vlasov equation [M. Tani *et al.*, *Phys. Rev. B* **104**, 075157 (2021)] to account for dynamic electron-electron scattering by introducing the Vlasov-Uehling-Uhlenbeck (VUU) equation. We further couple the VUU equation with Maxwell's equations to describe the laser pulse propagation. We apply the present approach to simulate laser-electron interactions in bulk and thin-film aluminum, focusing on energy absorption and transport. Our calculation results reveal that electron-electron scattering affects energy absorption more significantly under *p* polarization than under *s* polarization, highlighting the role of the nonuniform surface potential. Our simulations also show that the energy transport extends beyond the optical penetration depth, which is consistent with observations in previous laser ablation experiments. The developed Maxwell-VUU approach is expected to advance the understanding of intense laser-material interactions not only as a cost-effective alternative to the time-dependent density functional theory (TDDFT), but also by incorporating fermionic two-body collisions whose description is limited in TDDFT.

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I. INTRODUCTION

The interaction of ultrashort intense laser pulses with solids is relevant to various research fields ranging from high-harmonic generation [1–5] to material processing [6–14]. Ultrafast laser material processing, which suppresses heat-affected damage, starts from the energy transfer from the laser to the material by electron excitation, followed by instability of the lattice system or energy transfer from the heated electrons to the lattice. Next, the material undergoes phase and/or structural transition [15], leaving a change of the optical properties or a defect behind [16], which eventually leads to irreversible damage, drilling, or structuring [17–29].

The comprehensive modeling of laser material processing is highly complex, multiscale in both time and space, multiphase (solid, fluid, plasma, atomic gas, etc.), and possibly accompanied by chemical reactions. Plasma or continuum models [6,8,30–33] have been employed to describe and simulate the physical initial processes, advancing fundamental understanding. However, they have difficulties in examining most initial transient dynamics before the system reaches local thermodynamic equilibrium.

It has become possible to describe the nonequilibrium electron dynamics driven by strong electromagnetic waves using the time-dependent density-functional theory (TDDFT) [34–38] or time-dependent density-matrix methods [39–41]. TDDFT is a first-principles method that strikes a good balance between accuracy and computational feasibility. However, the computational cost is still very high for performing repeatedly to find optimal laser parameters. Another significant challenge with TDDFT is to find an exchange-correlation functional that can properly incorporate dynamical electronic correlations such as electron-electron scattering, though no systematic improvement exists.

Recently, we have developed a semiclassical method based on the Vlasov equation as a cost-effective alternative to

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TDDFT for studying electron dynamics in bulk solid metals under intense laser fields [42]. Starting from ground state distribution obtained by solving the Thomas-Fermi model, time evolution is calculated by pseudoparticle method. The optical spectra, including optical conductivity, dielectric function, complex refractive index, and reflectivity, are evaluated by linear response calculation using impulse response, and have been shown to agree well with TDDFT and experimental data. The absorbed energy, which has been evaluated as work done by laser field, has shown a linear dependence on the incident laser intensity. This trend has agreed with TDDFT results in a certain laser fluence region.

In this paper, we further extend the Vlasov-based approach [42] in two ways. First, we include dynamical electron-electron scattering, which is recognized as a vital process in metallic nanoclusters [43,44] and induces avalanche ionization in dielectrics and semiconductors in the context of laser material processing. Second, we consider the propagation effect of electromagnetic waves, which is crucial due to the fact that laser pulses typically used for laser machining can only penetrate the skin of metals.

To achieve the above purposes, in this work, we introduce the Vlasov-Uehling-Uhlenbeck (VUU) equation together with Maxwell's equations to describe coupled laser-electron dynamics in a metal thin film under intense laser fields. The VUU equation is an extension of the Vlasov equation taking into account fermionic two-body collision [45], which is difficult to treat with TDDFT. The VUU equation is numerically solved by pseudoparticle method [42], which represents the electron cloud as an assembly of classical pseudoparticles whose motion is governed by the Newton equation. The scattering term is evaluated by Monte Carlo sampling [43]. Then, we couple the VUU equation with Maxwell's equations, numerically solved with finite differential method. We apply the present method to a thin aluminum film. First, our VUU simulations reveal that the effect of electron-electron scattering on absorbed energy is significant for p -polarized light while it is minor for s polarization. Second, our Maxwell-VUU simulations show that the energy deposition reaches deeper than would be expected by laser pulse penetration alone.

The present paper is organized as follows. Section II describes our simulation methods. We review the VUU equation and Maxwell's equations, and describe our numerical implementations with the periodic boundary condition. In Sec. III we describe numerical application of VUU simulation and Maxwell-VUU simulation to bulk or thin aluminum. The conclusions are given in Sec. IV.

II. METHODS

A. The VUU equation

The VUU equation describes the dynamics of the electron distribution $f(\mathbf{r}, \mathbf{p}, t)$ in the phase space as

$$\frac{\partial}{\partial t} f(\mathbf{r}, \mathbf{p}, t) + \frac{\mathbf{p}}{m} \cdot \nabla_{\mathbf{r}} f(\mathbf{r}, \mathbf{p}, t) = \nabla_{\mathbf{r}} V_{\text{eff}}[n_e(\mathbf{r}, t)] \cdot \nabla_{\mathbf{p}} f(\mathbf{r}, \mathbf{p}, t) + I_{\text{UU}}(\mathbf{r}, \mathbf{p}, t), \quad (1)$$

where $\mathbf{r}$, $\mathbf{p}$, and m are the electron position, canonical momentum, and mass respectively, V_{eff} the effective mean-field

potential, and I_{UU} the two-body collision integral (see below) [43], which was absent in our previous work [42].

The effective potential V_{eff} is a functional of the electron density $n_e(\mathbf{r}, t)$ and decomposed into

$$V_{\text{eff}}[n_e(\mathbf{r}, t)] = V_{\text{Coulomb}}[n_e(\mathbf{r}, t)] + V_{\text{xc}}[n_e(\mathbf{r}, t)] + V_{\text{ext}}(\mathbf{r}, t), \quad (2)$$

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

$$V_{\text{Coulomb}}[n_e(\mathbf{r}, t)] = \sum_i V_{\text{ps}}(\mathbf{r} - \mathbf{r}_i) + V_H[n_e(\mathbf{r}, t)], \quad (3)$$

where i , V_{ps} , and V_H denote the label of ions and the spherically symmetric ionic pseudopotential and the electron-electron Hartree potential, respectively. In this work, we employ the modified Heine-Abarenkov local pseudopotential for V_{ps} [42,46]. V_H is evaluated by solving the Poisson equation,

$$\Delta V_H[n_e(\mathbf{r}, t)] = -4\pi e n_e(\mathbf{r}, t). \quad (4)$$

The laser-electron interaction is described in the length gauge,

$$V_{\text{ext}}(\mathbf{r}, t) = -e\mathbf{E}(t) \cdot \mathbf{r}, \quad (5)$$

within the dipole approximation, where $\mathbf{E}$ denotes the laser electric field vector. We use the local-density approximation (LDA) by Perdew and Zunger [47] for the exchange-correlation potential V_{xc} . Although the exchange-correlation potential in principle includes the effect of electron-electron scattering, it is known that this contribution is negligibly small within the LDA [48]. Therefore, there is little concern about double counting when combined with collision integrals.

In our previous study [42] we have solved the (collisionless) Vlasov equation or Eq. (1) with I_{UU} switched off, which can be derived as the leading order of a semiclassical $\hbar$ expansion of the time-dependent Kohn-Sham equation [42] and also be viewed as a dynamic generalization of the static Thomas-Fermi model. Although electron-electron scattering is, in principle, included in V_{xc} , it is significantly limited in practice. Therefore, we explicitly introduce the collision integral I_{UU} in Eq. (1).

To solve Eq. (1) numerically, we employ the pseudoparticle method, where the distribution $f(\mathbf{r}, \mathbf{p}, t)$ is treated as a set of many pseudoparticles:

$$f(\mathbf{r}, \mathbf{p}, t) \equiv \frac{1}{N_s} \sum_{i=1}^{N_{\text{pp}}} g_r[\mathbf{r} - \mathbf{r}_i(t)] g_p[\mathbf{p} - \mathbf{p}_i(t)]. \quad (6)$$

Here $\mathbf{r}_i$, $\mathbf{p}_i$ are the position and canonical momentum of each pseudoparticle labeled by i . The total number of pseudoparticles N_{pp} is expressed as $N_{\text{pp}} = N_s N_e$ where N_s, N_e are the number of pseudoparticles per electron and the total number of electrons contained in the simulation box Ω , respectively. N_s is set to 10 000 as it is sufficient to suppress statistical noise level [42]. g_r, g_p denote smoothing kernel functions in real and momentum space, respectively, of Gaussian forms,

$$g_r(\mathbf{r}) = \sum_{\mathbf{G}} \frac{1}{\pi^{3/2} d_r^3} \exp(-|\mathbf{r} + \mathbf{G}|^2/d_r^2), \quad (7)$$

$$g_p(\mathbf{p}) = \frac{1}{\pi^{3/2} d_p^3} \exp(-|\mathbf{p}|^2/d_p^2), \quad (8)$$

where d_r, d_p are smoothing widths in real and momentum space, respectively. Within the pseudoparticle method, the collision integral I_{UU} is treated as stochastic collision events among pseudoparticles taking place when accepted by Monte Carlo sampling [43]. At each time step, we randomly pick up a pair of pseudoparticles, whose momenta are $(\mathbf{p}_1, \mathbf{p}_2)$. If their distance is smaller than “impact parameter” $b = \sqrt{(\sigma_{\text{tot}}/\pi N_s)}$, we randomly choose a new momentum state after elastic collision $(\mathbf{p}_1, \mathbf{p}_2) \rightarrow (\mathbf{p}'_1, \mathbf{p}'_2)$, which is accepted by the rate $(1 - f_{\mathbf{p}'_1})(1 - f_{\mathbf{p}'_2})$ that accounts for Pauli blocking, where $f_{\mathbf{p}}$ denotes the local momentum occupation. Here σ_{tot} denotes the total scattering cross section that is derived as that by screened Coulomb potential which solely depends on the electron density (details are given in Ref. [43]). If the collision event mentioned above is not accepted, a new pair will be sampled, and the same process will be repeated. However, once a pseudoparticle is accepted in a scattering event, it will not be used for another scattering event within the same time step.

We employ a real-space simulation box Ω for the electron system, on which the periodic boundary condition is imposed [42]. Numerical implementation for the ground state calculation, described by the Thomas-Fermi model, and the time propagation for the left-hand side in Eq. (1) are described in Ref. [42] in details. The charge current density $\mathbf{J}(t)$ averaged over the simulation box Ω is given by

$$\mathbf{J}(t) = \frac{1}{|\Omega|} \iint_{\Omega} \left(-e \frac{\mathbf{p}}{m} \right) f(\mathbf{r}, \mathbf{p}, t) d\mathbf{r} d\mathbf{p}, \quad (9)$$

where $|\Omega|$ denotes the volume of the simulation box. The energy current is defined in an analogous way:

$$\mathbf{Q}(t) = \frac{1}{|\Omega|} \iint_{\Omega} \left(\frac{\mathbf{p}^2}{2m} \frac{\mathbf{p}}{m} \right) f(\mathbf{r}, \mathbf{p}, t) d\mathbf{r} d\mathbf{p}. \quad (10)$$

B. Maxwell's equations

We consider normal incidence of a linearly polarized laser pulse. The Maxwell equation to be solved is

$$\left(\frac{1}{c^2} \frac{\partial^2}{\partial t^2} - \Delta \right) \mathbf{A}(\mathbf{r}, t) + \frac{\partial}{\partial t} \nabla V_H(\mathbf{r}, t) = \mu_0 \mathbf{J}(\mathbf{r}, t), \quad (11)$$

where c denotes the light speed, $\mathbf{A}(\mathbf{r}, t) = -\int_{-\infty}^t \mathbf{E}(\mathbf{r}, \tau) d\tau$ the vector potential, $\mathbf{E}(\mathbf{r}, t)$ the electric field, and $\mathbf{J}(\mathbf{r}, t) = \int (-e \frac{\mathbf{p}}{m}) f(\mathbf{r}, \mathbf{p}, t) d\mathbf{p}$ the local current density at each grid point. Note that field quantities related to Eq. (11), $\mathbf{A}, \mathbf{E}, \mathbf{J}$, are of the Maxwell system. The VUU equation (1) and Maxwell equation (11) are coupled through the electric field and the current density. Here we follow the framework that has been used to couple the classical electrodynamics and the time-dependent density functional theory [49]. The electric field $\mathbf{E}(\mathbf{r}, t) = -\dot{\mathbf{A}}(\mathbf{r}, t)$, obtained from Eq. (11), is used as an input to Eq. (1). Pseudoparticles feel the electric field at the nearest grid point. The initial wave form in vacuum is given as a spatial distribution of the vector potential. At the boundary of the simulation box for the Maxwell equation, which is extended longer than Ω along the optical axis, Mur's absorbing boundary conditions are imposed to suppress unphysical reflection [50]. Figure 1 shows a schematic diagram in the x - y plane. While the VUU and Maxwell equations share the

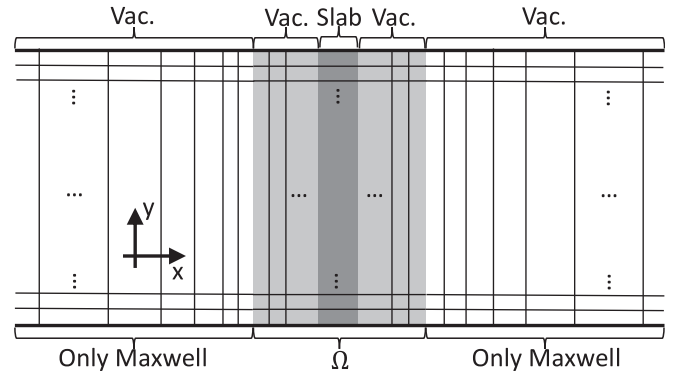


FIG. 1. Model geometry. Ω , which is composed of the slab region and the vacuum region, is the simulation box for both the electronic system and the Maxwell system. The vacuum domain outside Ω is only for the Maxwell system.

grid points inside Ω , the latter is solved also in vacuum layers attached to both sides of Ω .

III. RESULTS AND DISCUSSIONS

A. Linear response

Let us first examine the linear optical response by applying an impulse electric field which is equivalent to setting initial pseudoparticle momenta $\mathbf{p}_i = \mathbf{p}_i^{\text{GS}} + \Delta \mathbf{p}$ shifted from the ground state values $\mathbf{p}_i^{\text{GS}}$ by a small amount $\Delta \mathbf{p} = (0, 0, 0.1)$ a.u. [42]. In this simulation, we solely use the simulation box Ω on which the periodic boundary condition is applied for all the three directions with no vacuum region. Figure 2 shows the comparison of the calculated current density among the Vlasov and VUU simulations with the modified Heine-Abarenkov potential and a jellium model, where a uniform positive charge is used instead of ionic

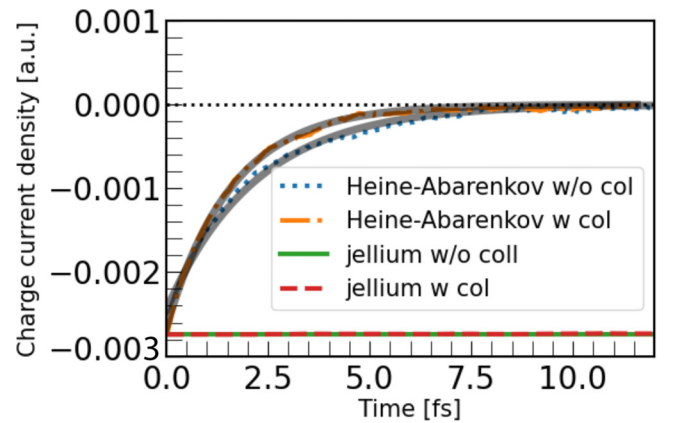


FIG. 2. Time evolution of charge current density after impulsively added momentum $\Delta \mathbf{p}$ at $t = 0$. Blue dotted and orange dash-dotted lines indicate Vlasov and VUU simulation results with the modified Heine-Abarenkov potential. Green solid and red dashed lines represent Vlasov and VUU simulation results with the jellium model. The black translucent lines represent the fitting results using Eq. (12) of the simulation curve with the Heine-Abarenkov type potential.

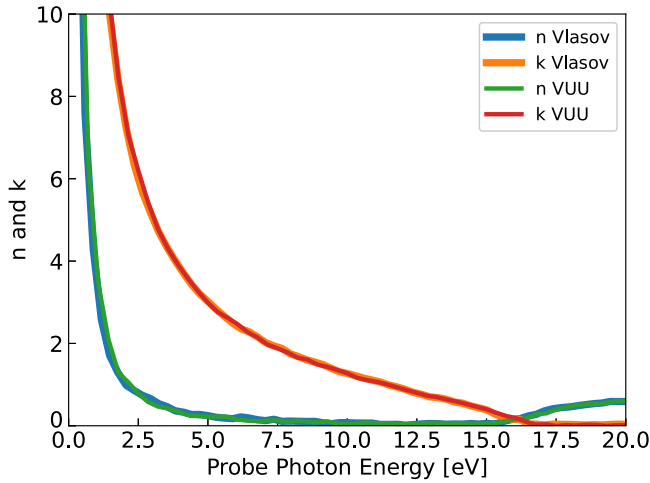


FIG. 3. Blue and orange solid lines denote n and k calculated by collisionless Vlasov simulation. Green and red are n and k calculated by VUU simulation.

pseudopotentials. The time evolution of the current density is fitted by

$$J_z(t) = J_z^0 \exp(-t/\tau), \quad (12)$$

where fitting parameters are the initial value J_z^0 and damping constant τ . τ is 2.2 and 1.6 fs for the collisionless (Vlasov) and the collisional (VUU) case, respectively. While both collisionless and collisional dynamics involve damping, the difference in τ implies that our simulations include momentum relaxation due to electron-electron scattering. It should be noted that the electron scattering from the ionic potential, which leads to the damping as is seen in Fig. 2, is a classical analog of Umklapp scattering [51]. In fact, in the calculation using the jellium model, we observe current damping for neither collisionless nor collisional calculations. Figure 3 compares the real and imaginary parts of the refractive index, n and k , between the collisionless and collisional simulations.

We see the effects of change in τ on local linear optical constants are quite small. The effects of the change in the relaxation time τ on the local linear optical constants are found to be very small. In the context of the Drude model, the quantity primarily affected by electron-electron scattering is the relaxation time τ . The impact of variations in τ on optical constants remains minor as long as the plasma frequency is sufficiently larger than $1/\tau$. This condition is satisfied in our simulations: the plasma frequency is approximately 0.5 a.u., whereas $1/\tau$ is about 0.01 a.u.

The number of pseudoparticles is 0.12×10^6 . Time evolution takes 0.06 s (collisionless) and 0.15 s (collisional) per time step using flat-MPI with 64 cores (using two Intel Xeon Platinum 8280 CPUs, attached 192GB DDR4 memory per each). Whole simulation takes 1.7 h (collisionless) and 4.3 h (collisional) with a time step 0.4838 as. For the collisionless simulation, time evolution takes 0.12 s per time step using flat-MPI with 96 cores (using two A64FX CPUs, attached 32 GB HBM2 memory per each). Using 243 A64FX CPUs, TDDFT requires 0.6838 h to propagate a primitive unit cell for 50 fs with the same time step. The reduction of computational time by our Vlasov simulation from TDDFT is estimated to be 1/100.

B. Energy transfer from laser to electrons

Let us next investigate the energy absorption of an aluminum film from the laser pulse. The electric field is assumed to be spatially uniform and given by

$$E(t) = E_0 \sin(\omega t) \cos^2 \left[\frac{t\pi}{T} \right] \left(-\frac{T}{2} \leq t \leq \frac{T}{2} \right), \quad (13)$$

where E_0 denotes the field amplitude, $\hbar\omega$ the photon energy, and T the (foot-to-foot) full pulse duration. The laser intensity profile has a full width at half maximum (FWHM) duration of approximately $0.36T$. In this subsection, Eq. (1) is solved without coupling with Eq. (11). We solely use simulation box Ω on which the periodic boundary condition is applied for the electron system. Ω contains a 4-nm vacuum region on both sides of the film along the optical axis.

Here we consider a central wavelength of 400 nm, peak intensity of 1 TW/cm^2 , and pulse width of 32.4 fs in FWHM (corresponding to 90 fs in foot-to-foot pulse width). This laser pulse is applied to a 4-nm-thick aluminum slab in a vacuum. The number of pseudoparticles is about 1.25×10^6 . Time evolution takes 0.27 s (collisionless) and 0.53 s (collisional) per time step using flat-MPI with 960 cores (corresponding to 40 A64FX CPUs). Whole simulation takes 14 h (collisionless) and 27 h (collisional) with a time step 0.4838 as.

We evaluate the absorbed energy as the difference in total electron energy from the initial state. Figure 4 shows the time evolution of the absorbed energy under s - and p -polarized laser pulses. Increase in absorbed energy due to electron-electron scattering is larger for p polarization ($\approx 2.5 \text{ eV}$) than for s polarization ($\approx 0.5 \text{ eV}$). This observation can be attributed to two factors: the scattering rate and the contribution from potential scattering. In the p -polarized case, the overall electron motion is perpendicular to the surface. The difference in the density of states between the surface and the bulk may lead to a temporally low occupancy, resulting in frequent electron-electron scattering due to the lower Pauli blocking rate. For the simulations with electron-electron scattering [blue solid line in Fig. 4(b)], the absorbed energy still increases during the trailing edge of the pulse, while it is constant after the pulse [inset of Fig. 4(b)]. A possible reason is that once the electrons are excited, the occupation in the momentum space becomes far from the step function, then electron-electron scattering occurs more easily after thermalization, leading to an increase in the absorption rate even under a weak laser field. A common process for both s polarization and p polarization is scattering from the ionic core potentials. However, the surface electrons are scattered by the surface Coulomb potential, further promoting collisional dissipation under p -polarized laser fields. It is easy to show that elastic collision of two particles at $t = t_{\text{col}}$ under a uniform external potential, upon which the momentum change is described by $(\mathbf{p}_1(t_{\text{col}}), \mathbf{p}_2(t_{\text{col}})) \rightarrow (\mathbf{p}'_1(t_{\text{col}}), \mathbf{p}'_2(t_{\text{col}}))$, does not change the final total energy:

$$\begin{aligned} & \frac{1}{2m}(\mathbf{p}_1 + \mathbf{A}_{\text{col}})^2 + \frac{1}{2m}(\mathbf{p}_2 + \mathbf{A}_{\text{col}})^2 \\ &= \frac{1}{2m}(\mathbf{p}'_1 + \mathbf{A}_{\text{col}})^2 + \frac{1}{2m}(\mathbf{p}'_2 + \mathbf{A}_{\text{col}})^2, \end{aligned} \quad (14)$$

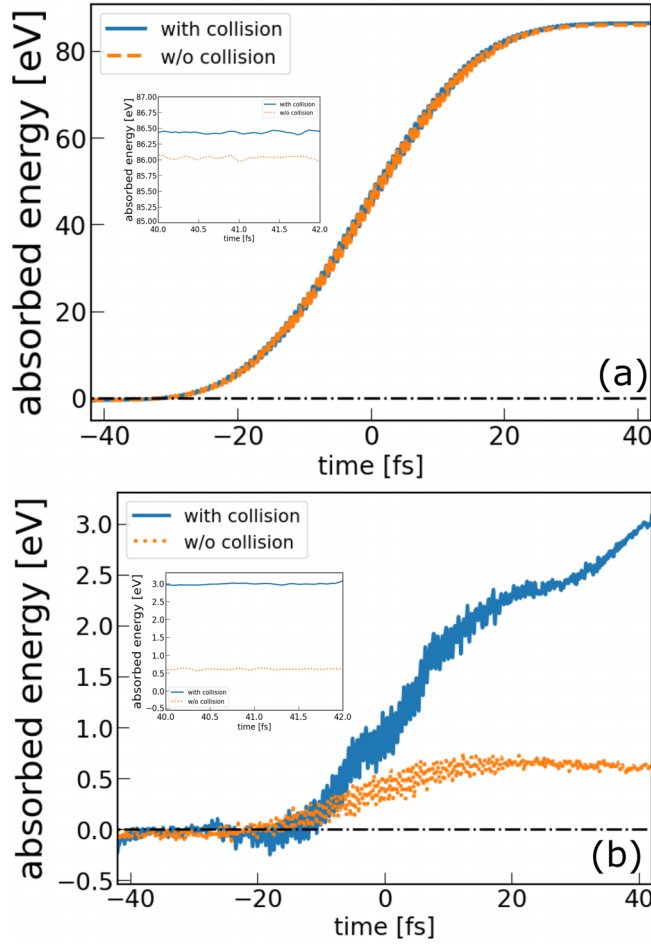


FIG. 4. Absorbed energy under (a) s -polarized light and (b) p -polarized light. Blue solid and orange dashed lines are results with and without electron-electron scattering. Insets are magnified views of the time period around the end of the laser pulse.

where $\mathbf{A}_{\text{col}}$ denotes the laser vector potential at the time of the collision. On the other hand, the acute nonuniformity of the surface Coulomb potential experienced by the returning electrons under p polarization leads to a larger increase in the final total energy.

The observed polarization-dependent energy absorption represents an anisotropic response that is beyond the common isotropic assumptions of the temperature models. Consistent with ultrafast experiments on metals, polarization-dependent effective penetration depth and ablation efficiency have been reported under femtosecond irradiation (e.g., Ref. [52]). The observation that the effective energy deposition extends well beyond the optical skin depth in a polarization-dependent manner cannot be explained by a purely diffusive energy transport picture assumed in the two-temperature model, and instead indicates the presence of a nondiffusive, at least partly ballistic, electron energy transport component. In our framework, such polarization dependence arises naturally from the combined action of the mean-field (electron-ion/electron-electron) forces and explicit electron-electron collisions (UU), providing a mechanistic basis for the macroscopic anisotropy. More generally, the Vlasov-VUU framework continuously interpolates between the ballistic and diffusive transport

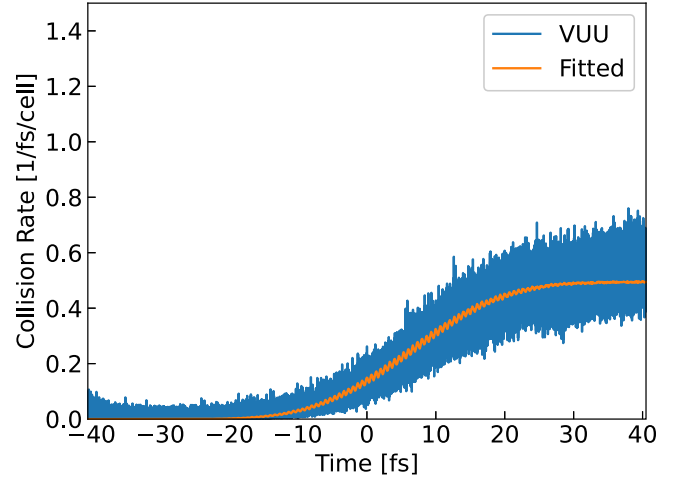


FIG. 5. Blue and orange lines indicate the electron-electron scattering rate σ_{ee} of the VUU simulation with s polarization and the fitted curve.

regimes: when electron-electron scattering is sufficiently frequent, the ballistic contribution is effectively suppressed and the energy transport approaches a diffusion-dominated behavior, consistent with temperature-based models.

We now examine the validity of the electron-electron scattering rate in our formalism, σ_{ee} , defined as the number of scattered electrons per unit time over a whole cell. A direct quantitative comparison with experiments is challenging because σ_{ee} is difficult to measure directly. Instead, we test the expected quadratic dependence of σ_{ee} on the electron temperature T_e [53]. Figure 5 shows the real-time evolution of σ_{ee} together with a fitting curve based on $\sigma_{ee} \propto K_e^2$, where K_e denotes the electron kinetic energy. The fitting curve reproduces the real-time data well, indicating that our formalism provides a reasonable description of electron-electron scattering.

C. Energy transport

In this subsection, we utilize the Maxwell-Vlasov-(UU) simulation to investigate how the electrons are excited in an aluminum film which has 16-nm thickness under a propagating linearly polarized laser pulse whose initial electric field profile is given by

$$E(x, t = 0) = E_0 \sin\left[\frac{\omega}{c}x\right] \sin^2\left(\frac{x}{cT}\pi\right) \quad (0 \leq x \leq cT). \quad (15)$$

We use simulation box Ω on which the periodic boundary condition is applied for the electron system. Ω contains 4-nm vacuum regions on both sides of the film along the optical axis. The simulation box for the Maxwell equation has additional vacuum layers of 4- μm thickness attached to both sides of Ω .

We consider a laser pulse with a central wavelength of 200 nm, peak intensity of 100 TW/cm^2 , and FWHM pulse width of 3.6 fs (10 fs foot-to-foot), normally incident to a 16-nm-thick aluminum slab in a vacuum. Figure 6 shows the temporal evolution of the total, kinetic, and potential energies for collisional and collisionless simulations. The total energy is the sum of the kinetic and the potential energies. We can

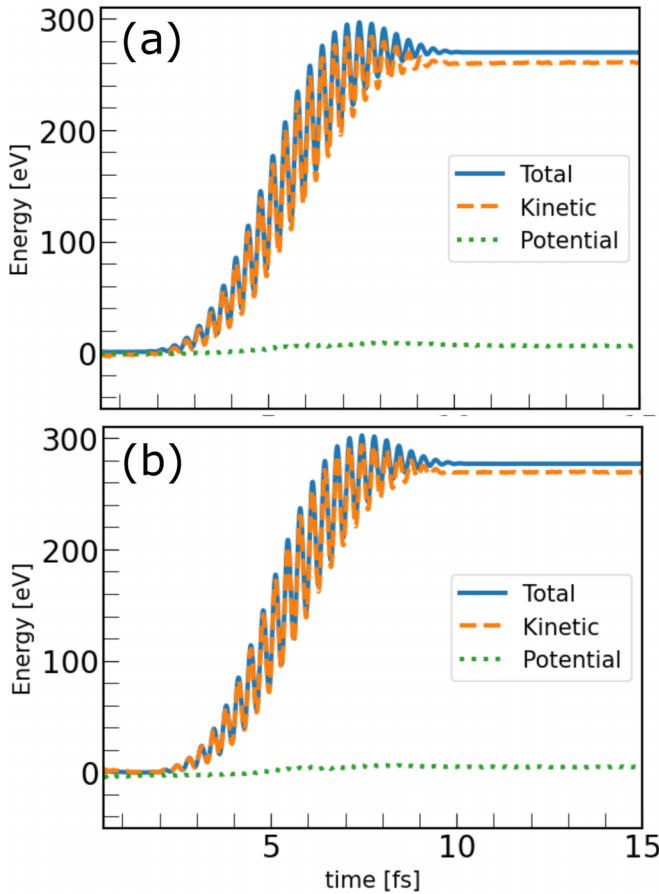


FIG. 6. Time evolution of energy gain is illustrated. Total energy is plotted in solid blue line, kinetic energy in dashed orange line, and potential energy in dotted green line. (a) Without collision. (b) With collision.

see that the kinetic energy gain is dominant for both collisional and collisionless cases, and, thus, let us focus on its dynamics. Figure 7 presents the depth distribution of the kinetic energy just after irradiation. The normalized laser peak intensity at each position along the optical axis is also plotted. The kinetic energy distribution is not proportional to the depth profile of the laser intensity but penetrates deeper in the sample, which means energy transport along the optical axis is taking place during a short time of laser pulse irradiation. These microscopic differences in energy flow have direct macroscopic consequences. In particular, they affect the effective electron penetration depth and ablation thresholds/efficiency [52]. The deeper energy delivery observed here, beyond the optical penetration depth, is expected to manifest itself as a larger effective penetration depth and modified thresholds under ultrafast excitation of metal thin films. The difference between the optical and electron heat penetration depths has been reported experimentally by Miyasaka *et al.* [52]. Our results support such observations based on a microscopic theoretical framework. Let us next quantitatively explore the energy flow given by Eq. (10) and how electron-electron scattering affects its behavior. Figure 8 shows the temporal evolution of the energy current density averaged on the whole space including the vacuum domains. The energy flow exists only along the

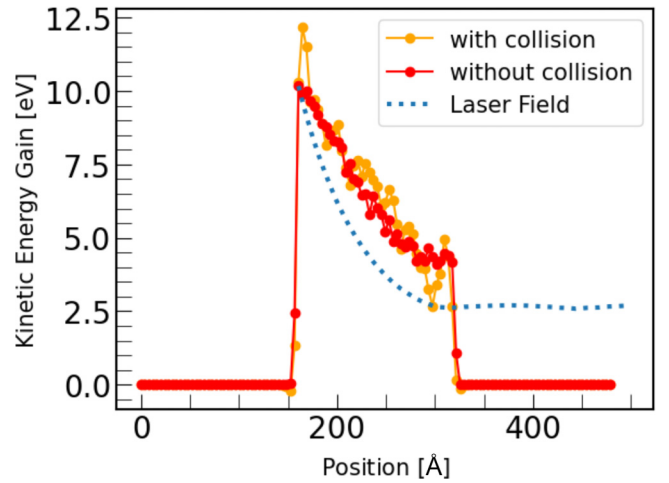


FIG. 7. Kinetic energy gain distribution calculated by collisionless simulation in red line and by collisional simulation in orange, and scaled maximum laser field intensity along optical axis in blue dashed line.

optical axis (x direction), with negligible net lateral components. In addition, we see that the energy current along the optical axis is reduced by electron-electron scattering, which partially redirects the electron motion to transverse directions.

The suppression of energy conduction in metal electrons due to electron-electron scattering is a well-known phenomenon in thermally equilibrated electron systems [54,55]. The computational results in this study demonstrate that a similar effect occurs in the nonequilibrium state induced by laser irradiation in metals. Despite this observation, the distribution of the kinetic energy gain in Fig. 7 is surprisingly similar for the collisional and collisionless cases. Indeed, the “center of mass” (c.m.) of the distribution with collision (225.1 Å) is only slightly deeper than that without collision (223.3 Å). On the other hand, c.m. of the distribution of the total energy gain

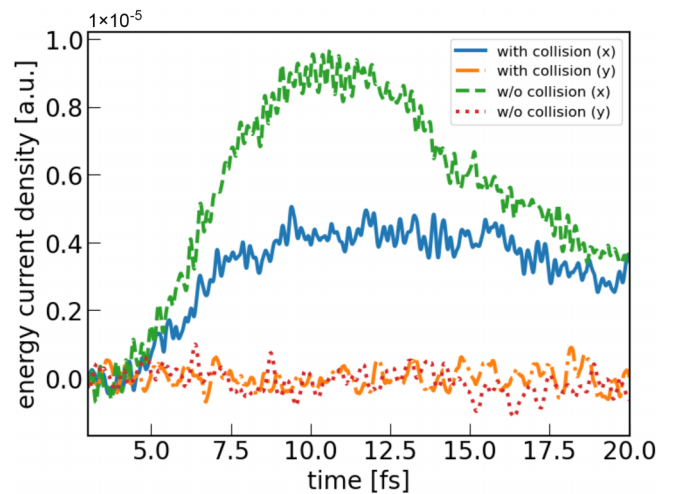


FIG. 8. Time evolution of energy current density in x and y direction filtered by moving average to reduce noise. Blue solid and green dashed lines indicate x components with and without electron-electron scattering; orange dash-dotted and red dotted lines indicate y components with and without electron-electron scattering.

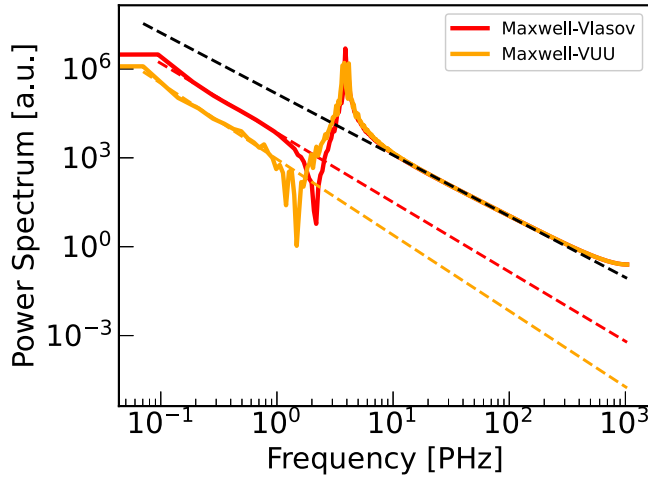


FIG. 9. Power spectrum of autocorrelation of the x component of energy flow. Red and orange solid lines indicate the spectrum. Dashed lines of corresponding colors are least-square fitting from 0.1 to 1 PHz; slopes of $\nu^{-2.3}$ and $\nu^{-2.6}$ are shown by red and orange lines. The black dashed line is fitted from 10 to 100 PHz, with slope of ν^{-2} .

gets deeper from 298.5 to 318.7 Å, which may suggest that the kinetic energy is eventually transformed to the potential energy.

We note that ballistic transport is inherently included in our framework: the Vlasov dynamics describe collisionless propagation under the coherent scattering field in the mean-field level, while the UU term provides hard electron-electron collisions, so that a ballistic-to-diffusive crossover is expected to emerge from the dynamics without prescribing transport coefficients [56]. Figure 9 shows the power spectrum of normalized biased autocorrelation of the x component of energy flow after reaching peak intensity ($t > 10$ fs) $S(\nu) = \mathcal{F}[A(\tau)/A(0)]$, where $A(\tau) = \frac{1}{T} \int_0^{T-\tau} \tilde{Q}_x(t) \tilde{Q}_x(t + \tau) dt$, $\tilde{Q}_x(t) = Q_x(t) - \bar{Q}$, $\bar{Q} = \frac{1}{T} \int_0^T Q_x(t) dt$, and ν stands for the frequency.

The power spectra of the energy-current autocorrelation exhibit a prominent resonance (collective mode) and interference minima at a few petahertz, and a power-law high-frequency tail. Compared with Maxwell-Vlasov, the Maxwell-VUU approach systematically suppresses the spectral amplitude and broadens the resonance, below ≈ 10 PHz, indicating reduced coherence and shorter correlation time, while preserving the high-frequency scaling exponent. Performing least-square fitting, the high frequency part of $10 \leq \nu \leq 100$ PHz is estimated to have a slope of ν^{-2} regardless of the UU term, indicating electron-electron collision does not affect ballistic energy flow within 0.1-fs scale or faster. In addition, the middle part of $0.1 \leq \nu \leq 1$ PHz is estimated to have slopes of $\nu^{-2.3}$ and $\nu^{-2.6}$ for Vlasov and VUU simulations, respectively, indicating a time-dependent relaxation rate of the energy flow at a few femtosecond scale. While long-time/thick-film runs are currently cost limited, this analysis already indicates a ballistic-to-diffusive transitioning tendency. Further code optimization and increased computing resources will extend these comparisons.

Let us discuss how the modification in the exchange-correlation term and two-body collisions affect the contribution of the ballistic transport. The average kinetic energy per electron is a few eV, which is lower by an order of magnitude than the Fermi temperature T_F . Under this condition, the inverse lifetime of electronic ballistic flux is known to be $1/\tau_e \propto 1/\tau_{ei} + 1/\tau_{ee}$ [55], where $\tau_{ee}(\propto 1/T_e^2)$ and τ_{ei} denote electron scattering contribution by electrons and ions, respectively. However, since the exchange-correlation term V_{xc} includes little contribution from electron-electron collisions, the previous Vlasov approach largely ignores the term $1/\tau_{ee}$, leading to $1/\tau_e \propto 1/\tau_{ei}$. On the other hand, in the VUU approach, which includes the two-body collision through I_{UU} , the T_e^2 contribution is properly taken into account. This effectively reduces τ_e , which is consistent with Fig. 2.

IV. CONCLUSIONS

We have extended the semiclassical formalism based on the Vlasov equation [42] to incorporate electron-electron scattering as well as propagating laser field dynamics. The electronic distribution function is expressed by a set of pseudoparticles. Electron-electron scattering is treated as collision events between pseudoparticles, where the Pauli blocking factor is taken into account in the acceptance rate. The propagation of the laser field is calculated by solving the Maxwell equations.

We have applied the developed approach to crystalline and slab aluminum. After the application of an impulse electric field to bulk aluminum, the VUU calculation leads to a faster damping of the charge current than the collisionless Vlasov calculation, due to the momentum relaxation by interelectronic scattering. The investigation of the effect of the laser polarization on energy absorption in an aluminum film has shown that electron-electron scattering increases the absorbed energy, especially under p polarization, rather than under s polarization. This result indicates an important role of the nonuniform surface Coulomb potential. Furthermore, our Maxwell-Vlasov(-UU) simulations have demonstrated that energy is transported deeper into the aluminum film than would be expected from the laser penetration depth alone. Although electron-electron scattering suppresses the energy flow along the optical axis, it does not significantly alter the spatial distribution of the kinetic energy gain, implying the transformation of the kinetic energy into potential energy. The linear optical constants remain essentially unaffected by including electron-electron scattering. In contrast, the strong-field response exhibits drastic changes in energy flow and energy absorption. The microscopic energy-flow changes reported here provide a mechanistic route to larger electron penetration depth and modified ablation thresholds/efficiency under ultrafast excitation, while the underlying complex relaxation processes arise self-consistently within one framework.

These extensions allow the investigation of electron dynamics in bulk and slab metals under ultrashort intense laser pulses while incorporating the effects of electron-electron scattering as well as laser pulse propagation, going beyond macroscopic modeling. The former extension offers an advantage over TDDFT, since the description of electron-electron collisions in TDDFT is limited. The latter extension overcomes the limitation of our previous Vlasov simulator, as laser

propagation becomes potentially important when the object size is not sufficiently smaller than the laser wavelength or when it is comparable to characteristic electromagnetic length scales. The next step will be to incorporate molecular dynamics, which plays a crucial role after strong electron excitation, leading to laser-induced irreversible damage.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX: FINITE WIDTH PSEUDOPARTICLES

One should carefully derive representations of moments using one-body distribution approximated by the pseudoparticle method. Assuming a two-dimensional classical phase space, the first-order moment $A(t)$ and second-order moment $B(t)$ are defined as

$$A(t) = \int_{\Gamma} \boldsymbol{\gamma} f_D(\mathbf{r}, \mathbf{p}, t) d\mathbf{r} d\mathbf{p}, \quad (\text{A1})$$

$$B(t) = \int_{\Gamma} \boldsymbol{\gamma}^2 f_D(\mathbf{r}, \mathbf{p}, t) d\mathbf{r} d\mathbf{p}, \quad (\text{A2})$$

where $\boldsymbol{\gamma} = \mathbf{r}, \mathbf{p}$, f_D stands for a N_p -physical-particle one-body distribution, and

$$\int_{\Gamma} f_D(\mathbf{r}, \mathbf{p}, t) d\mathbf{r} d\mathbf{p} = N_p. \quad (\text{A3})$$

Under the pseudoparticle ansatz, one-body distribution f_D is represented as a function of Gaussian kernels:

$$f_D(\mathbf{r}, \mathbf{p}, t) = \frac{1}{N_s} \sum_{i=1}^{N_{pp}} \frac{1}{(\sqrt{\pi} d_{\gamma})^{2D}} \times \prod_{\boldsymbol{\gamma}=\mathbf{r},\mathbf{p}} \exp \left[-\left(\frac{\boldsymbol{\gamma} - \boldsymbol{\gamma}_i(t)}{d_{\gamma}} \right)^2 \right], \quad (\text{A4})$$

where $N_s = N_{pp}/N_p$ denotes the number of pseudoparticles per physical particle. Using the representation in Eq. (A4), $A(t)$, $B(t)$ are written as

$$A(t) = \frac{1}{N_s} \sum_{i=1}^{N_{pp}} A_i(t), \quad (\text{A5})$$

$$A_i(t) = \int_{\Gamma} \boldsymbol{\gamma} \frac{1}{(\sqrt{\pi} d_{\gamma})^D} \exp \left[-\left(\frac{\boldsymbol{\gamma} - \boldsymbol{\gamma}_i(t)}{d_{\gamma}} \right)^2 \right] d\boldsymbol{\gamma}, \quad (\text{A6})$$

$$B(t) = \frac{1}{N_s} \sum_{i=1}^{N_{pp}} B_i(t), \quad (\text{A7})$$

$$B_i(t) = \int_{\Gamma} \boldsymbol{\gamma}^2 \frac{1}{(\sqrt{\pi} d_{\gamma})^D} \exp \left[-\left(\frac{\boldsymbol{\gamma} - \boldsymbol{\gamma}_i(t)}{d_{\gamma}} \right)^2 \right] d\boldsymbol{\gamma}. \quad (\text{A8})$$

$A_i(t)$, $B_i(t)$ are analytically evaluated as

$$A_i(t) = \boldsymbol{\gamma}_i(t), \quad (\text{A9})$$

$$B_i(t) = \boldsymbol{\gamma}_i(t)^2 + \frac{D d_{\gamma}^2}{2}, \quad (\text{A10})$$

therefore,

$$A(t) = \frac{1}{N_s} \sum_{i=1}^{N_{pp}} \boldsymbol{\gamma}_i(t), \quad (\text{A11})$$

$$B(t) = \frac{1}{N_s} \sum_{i=1}^{N_{pp}} \left[\boldsymbol{\gamma}_i(t)^2 + \frac{D d_{\gamma}^2}{2} \right] \quad (\text{A12})$$

$$= \frac{1}{N_s} \sum_{i=1}^{N_{pp}} \boldsymbol{\gamma}_i(t)^2 + \frac{D d_{\gamma}^2}{2} N_p. \quad (\text{A13})$$

When one employs the delta function as a kernel function on the $\boldsymbol{\gamma}$ space, or $d_{\gamma} = 0$, second-order moment $B^{\delta}(t)$ is defined as

$$B^{\delta}(t) = \frac{1}{N_s} \sum_{i=1}^{N_{pp}} B_i^{\delta}(t), \quad (\text{A14})$$

$$B_i^{\delta}(t) = \int_{\Gamma} \boldsymbol{\gamma}^2 \delta[\boldsymbol{\gamma} - \boldsymbol{\gamma}_i(t)] d\boldsymbol{\gamma} \quad (\text{A15})$$

$$= \boldsymbol{\gamma}_i(t)^2. \quad (\text{A16})$$

Inserting Eqs. (A14)–(A16) into Eq. (A13), one obtains

$$B(t) = B^{\delta}(t) + \frac{D d_{\gamma}^2}{2} N_p. \quad (\text{A17})$$

On a N_{pp} -pseudoparticles simulation, the statistical mean square error of the first term in Eq. (A17) would be order $\mathcal{O}(1)$ ($N_s \rightarrow \infty$). Although the second term is not necessarily negligible if one sets a finite d_{γ} , it is just a constant shift of energy. Even if d_{γ} is not zero, finite d_p correction brings essentially no change to our results from physical context.

Actually, using a finite d_p , one can evaluate kinetic energy $K(t)$ of the N_e electrons whose mass is m as

$$K(t) = \int_{\Gamma} \frac{\mathbf{p}^2}{2m} f_3(\mathbf{r}, \mathbf{p}, t) d\mathbf{r} d\mathbf{p} \quad (\text{A18})$$

$$= \frac{1}{2m} \left[\frac{1}{N_s} \sum_{i=1}^{N_{pp}} \mathbf{p}_i(t)^2 + \frac{3d_p^2}{2} N_e \right], \quad (\text{A19})$$

in a six-dimensional phase space, $D = 3$.

However, the second term in Eq. (A17) could not be missed when the absolute value of the electron energy plays an important role. For instance, heat current $J_h(t)$ is defined as

$$J_h(t) = \int_{\Gamma} \frac{\mathbf{p}^2}{2m} \frac{\mathbf{p}}{m} f_3(\mathbf{r}, \mathbf{p}, t) d\mathbf{r} d\mathbf{p} \quad (\text{A20})$$

$$= \frac{1}{N_s} \sum_i \frac{\mathbf{p}_i(t)^2}{2m} \frac{\mathbf{p}_i(t)}{m}. \quad (\text{A21})$$

One would see a non-negligible role of the finite d_p correction, unless $d_p \ll p$ for a typical absolute value of the kinetic momentum p .

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