High performance wave packet molecular dynamics with density functional exchange-correlation term for non-ideal plasma simulations

Ya S Lavrinenko¹,², I V Morozov¹,² and I A Valuev¹
¹ Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg 2, Moscow 125412, Russia
² Moscow Institute of Physics and Technology, Institutskiy Pereulok 9, Dolgoprudny, Moscow Region 141701, Russia
E-mail: lavrinenko@phystech.edu

Abstract. We report on development of the wave packet molecular dynamics (WPMD) with density functional theory (DFT) simulation technique that we proposed earlier for nonideal plasma and warm dense matter simulations. The method is based on the WPMD where the electron exchange-correlation effects are taken into account using the DFT approach. It is aimed at studying simultaneous dynamics of electrons and ions in equilibrium and non-equilibrium conditions for a wide range of temperatures and densities. Compared to classical molecular dynamics and WPMD simulations the method of WPMD-DFT provides more accurate representation of quantum effects such as electron–ion coupling and electron degeneracy. At this stage of the method development we pay a special attention to the performance issues such as acceleration with graphical processing units, the choice of an optimal simulation box size with respect to the boundary conditions, the use of an adaptive mesh for calculation of the exchange-correlation energy, and implementation of our algorithm in the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS). The results for internal energy of equilibrium dense hydrogen plasma are presented for evaluation of the method.

1. Introduction
A combination of the wave packet molecular dynamics (WPMD) with density functional theory (DFT) simulation technique that we proposed earlier [1]. The area of applicability of this new method includes equilibrium properties and dynamical (non-equilibrium) processes in electron–ion nonideal (strongly coupled) plasmas and warm dense matter such as equation of state, electron–ion relaxation, electron and ion plasma waves, plasma conductivity, reflectivity, diffusion, etc. These properties are of importance in view of recent experiments with shock-compressed gases [2, 3], laser ablation [4, 5], interaction of femtosecond laser pulses with nanoparticles [6-8] and so on.

Primarily, the WPMD-DFT method is based on the WPMD approach [9-12], which is an extension of classical molecular dynamics (MD). Here and below by the classical MD we mean solution of the Newtonian equations of motion for electrons and ions both represented by point-like particles [8, 13-16]. One should distinguish this method from the classical MD of atoms where the electrons are not taken into account directly. The later is widely used to study liquids and solids.
We follow the main idea of WPMD to represent an electron wave function as a Gaussian wave packet (WP) where the variable parameters are the WP position and width. There are few branches in development of WPMD-like methods such as electron force field (eFF) [17], antisymmetrized or Hartree–Fock WPMD (AWPMD) [18–20], WPMD with wave packet splitting [21, 22]. These modifications have some advantages but usually at the expense of performance. We use the original formulation of the WPMD approach [9] with a single wave packet per electron and the use of Hartree (non-antisymmetrized) approximation for the many-electron wave function. At the same time we include an additional term in the system Hamiltonian to account for electron exchange-correlation effects. This term is calculated as a functional of the local electron density following the DFT approach. As calculation of the exchange-correlation energy is performed on a spatial mesh, it could influence the simulation time significantly. Thus optimization of these calculations is critical for the overall method performance. Indirectly it affects also the accuracy of the method as a better performance allows using more particles in the simulation box and performing more averaging over a single simulation trajectory or an ensemble of independent trajectories.

Further performance gains can be achieved by executing code in parallel on a supercomputer. Parallel algorithms for molecular dynamics have been developed greatly in the last two decades, thus we decided to use a well known Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) package [23] as a basis for the parallel version of the WPMD-DFT code. The LAMMPS is known for its outstanding scalability and parallel efficiency for ordinary molecular dynamics. Moreover implementation of our algorithm in LAMMPS give such benefits as a ready to use pre- and post-processing tools, input script, output formats compatible with visualization and other packages.

Another feature of the WPMD-DFT code is the use of reflecting boundary conditions [1, 24] instead of periodic boundaries typically used in molecular dynamics simulations. It solves the problem of infinite wave packet spreading for weakly coupled electrons [12]. Below we analyze the influence of these boundaries and the simulation box size on simulation results for equilibrium nonideal plasma.

The wave packet method allows creating two types of algorithms: molecular dynamics for obtaining a time evolution of the system (WPMD) and Monte-Carlo for studying equilibrium systems using the Metropolis algorithm (WPMC). In this paper we report on simulations of equilibrium states only so that we mainly use the Monte-Carlo version which is similar to the molecular dynamics from the computational point of view; the only difference is calculation of the total energy derivatives which is relevant only for the dynamics simulations.

2. Simulation methods
2.1. Wave packet molecular dynamics
The details of WPMD method are described in [10, 19, 20], here we briefly mention their main principles. In these methods a single electron wave function is represented as a normalized Gaussian wave packet [9] described by a set of eight scalar parameters including the WP position \( \mathbf{r} \), width \( s \) and their conjugate momenta \( \mathbf{p} \), \( \mathbf{p}_s \):

\[
\varphi(\mathbf{x}) = \left( \frac{3}{2\pi s^2} \right)^{3/4} \exp \left\{ - \left( \frac{3}{4s^2} - \frac{ip_s}{2\hbar s} \right) (\mathbf{x} - \mathbf{r})^2 + \frac{i}{\hbar} \mathbf{p} \cdot (\mathbf{x} - \mathbf{r}) \right\}. \tag{1}
\]

Within the Hartree approximation a many-body wave function is given as a product of the single electron wave functions

\[
\Psi(\{\mathbf{x}_k\}) = \prod_{k=1}^{N_e} \varphi(\mathbf{x}_k), \tag{2}
\]
where \( N_e \) is the number of electrons. The resulting equations of motion for the parameters of all wave packets follow from the variational principle. They resemble the classical equations of motion in the fact that they may be derived from the generalized Hamiltonian function, which is the quantum expectation value of the system energy:

\[
H = \langle \Psi | \hat{H} | \Psi \rangle ,
\]

\[
\hat{H} = \sum_{k=1}^{N_e} \left( \frac{\hat{p}_k^2}{2m_e} + \frac{\hat{p}_{sk}^2}{2m_e} + \frac{9\hbar^2}{8m_es_k^2} - E_{0k} \right) + U_{ee}^{\text{wpmd}} + U_{ei}^{\text{wpmd}} + U_{ext} + K_i + U_{ii},
\]

where \( m_e \) and \( e \) are the mass and charge of the electron, \( Z_j \) and \( \mathbf{R}_j \) are the charges and positions of ions, \( N_i \) is the number of ions, \( U_{ext} \) is an external energy related usually to the boundary conditions, \( K_i \) and \( U_{ii} \) are the kinetic energy of ions and the energy of ion–ion interaction.

Analogously to simulations of classical systems, the equilibrium properties of the WPMD system may be obtained either by solving the equations of motion (temporal evolution) or by methods. Here we briefly list these issues and specify how we address them in the present work.

Let us rewrite the effective Hamiltonian function (3) as

\[
H = \sum_{k=1}^{N_e} \left( \frac{\hat{p}_k^2}{2m_e} + \frac{\hat{p}_{sk}^2}{2m_e} + \frac{9\hbar^2}{8m_es_k^2} - E_{0k} \right) + U_{ei}^{\text{wpmd}} + U_{ee}^{\text{wpmd}} + U_{ext} + K_i + U_{ii},
\]

where the effective potential energies are

\[
U_{ei}^{\text{wpmd}} + U_{ee}^{\text{wpmd}} + U_{ext} = \sum_{k=1}^{N_e} \left[ E_{0k} + \sum_{l=1}^{N_e} U_{\text{wall}}(\mathbf{r}_k) - \sum_{j=1}^{N_i} \frac{e^2 Z_j}{|\mathbf{r}_k - \mathbf{R}_j|} \text{erf} \left( \frac{\sqrt{3}|\mathbf{r}_k - \mathbf{R}_j|}{\sqrt{2}s_k} \right) \right] + \sum_{m<k} \frac{e^2}{|\mathbf{r}_k - \mathbf{r}_m|} \text{erf} \left( \frac{\sqrt{3}|\mathbf{r}_k - \mathbf{r}_m|}{\sqrt{2}(s_k^2 + s_m^2)^{1/2}} \right).
\]

In the Hartree case, the kinetic and potential energies are decoupled, where “kinetic energy” denotes here the expectation value of the first two terms in the sum in (4). The quantity \( E_{0k} \) is the energy of the equilibrium state for the width of each electron. For bound electrons this state corresponds to the electron localized at the ionic centre, and for “free” or extended electrons it is determined either by Pauli repulsion (antisymmetrized case) or by the confinement walls. Assuming that in the thermally equilibrated plasma the mean contributions from the kinetic energy \( \langle p_{sk}^2/2m_e \rangle \) and the “width potential” \( (9\hbar^2/(8m_es_k^2) - E_0) \) equal to \((1/2)N_e kT\) we end up with the total excess energy of \( N_e kT \) which should be subtracted from the total energy of the WPMD system for proper comparison with the classical MD results.

For the WPMC simulations, we use the Monte-Carlo sampling with variations in all coupled degrees of freedom, including ion positions, wave packet centres and momenta, width and width momenta to equilibrate the system at the given temperature \( T \). Adaptive steps are used to keep the mean Monte Carlo acceptance rate at 0.5. The ionic kinetic energy is decoupled and assumed to be equal to \((3/2)N_i k_B T\). The correction of \( N_e k_B T \) for extra degrees of freedom is subtracted from the obtained equilibrium energies.

2.2. Boundary conditions and calculation of pressure

One of the major problems of using the original WPMD method for simulations of spatially unlimited systems is the unlimited broadening of the Gaussian wave packets. In our earlier
work [24] we suggested to use a 3-dimensional confining potential for limiting the wave packet widths

$$U_{\text{wall}}(x) = \begin{cases} 
  k(|x| - L/2)^2, & |x| > L/2, \\
  0, & |x| \leq L/2,
\end{cases}$$

(6)

where $L = n_e^{-1/3}N_e$ is the length of the simulation box edge. The parameter $k$ determines the strength of the reflecting potential, acting on the wave packets and classical ions. The value of $k$ is selected large enough to keep the particles mostly inside the MD cell. However, too large values of $k$ may lead to large forces and destabilize the time stepping. Unlike other limiting approaches [12], this approach does not introduce any additional non-physical restrictions on the wave packet widths.

In the classical MD with periodic boundary conditions the following virial expression is used for evaluating the pressure

$$P = \frac{2}{3V} \sum_{i=1}^{N} m_i v_i^2 - \frac{1}{3V} \sum_{i,j=1}^{N} r_{ij} \frac{\partial U(r_{ij})}{\partial r_{ij}},$$

(7)

where $V$ is the system volume, $m_i$ and $v_i$ are mass and velocity of the $i$-th particle, $r_{ij}$ is the interparticle distance [25]. The use of expression (7) in the WPMD method with the confining potential may be incorrect, since formally a transition from the virial theorem to expression (7) is not valid because of the existence of external forces. Although the use of (7) is possible in the internal region not affected by the confinement. In the present work we rely on the alternative pressure evaluation approach outlined below.

With the reflecting boundary conditions the pressure can be obtained directly by evaluating the confinement forces $F_{\text{wall}}^\alpha$ acting on particles along an axis $\alpha$

$$P = \frac{1}{3} \left\langle P_x + P_y + P_z \right\rangle = \frac{1}{3} \left\langle \sum_{\alpha=x,y,z} \frac{|F_{\text{wall}}^\alpha|}{S_{\alpha}} \right\rangle,$$

(8)

where $S_{\alpha}$ is the area of the corresponding simulation box side.

2.3. Extending wave packet molecular dynamics using the density functional theory

The original WPMD method with the Hartree approximation can be extended to account for exchange and correlation effects by introducing an additional energy term $E_a$ calculated from the electron density with the use of an exchange-correlation functional (see [1])

$$\hat{H}_{\text{wpmd–dft}} = \hat{H}_{\text{wpmd}} + E_a[n],$$

(9)

where $n = n(r)$ is the total electron density. In WPMD the density can be evaluated as a sum of the wave packet norms

$$n(r) = \sum_{k=1}^{N_e} \varphi_k(r)\varphi_k^*(r).$$

(10)

The expression for $E_a$ contains terms that describe exchange and correlation energy in the local density approximation (LDA) $E_{XC}[n]$ and additional terms for compensating the self-interaction $E_{\text{self}}$ and correcting the kinetic energy for Pauli degeneration $T_s[n] - T_{WP}$

$$E_a[n] = T_s[n] - T_{WP} + E_{XC}[n] + E_{\text{self}}.$$

(11)

Following the Hartree–Fock WPMD [19] we may assign constant spin values to the wave packets so that a half of them has spin up and another half has spin down. Then the local spin density approximation (LSDA) can be applied

$$E^{\text{LSDA}}_{XC}[n_\uparrow, n_\downarrow] = \int \varepsilon_{XC}(n_\uparrow, n_\downarrow)n(r) \, dr, \quad n(r) = n_\uparrow(r) + n_\downarrow(r),$$

(12)
\[ T_s[n] = \frac{3}{10} (3\pi^2)^{2/3} \int n(r)^{5/3} \, dr. \] (13)

Calculation of the exchange and correlation energies with respect to the wave packet parameters is performed by an evaluation of functional (11) on a spatial mesh

\[ E_a[n] = \sum_{i=1}^{M_x} \sum_{j=1}^{M_y} \sum_{k=1}^{M_z} E_{a,ijk}[n(r_{ijk})], \] (14)

\[ E_{a,ijk}[n(r_{ijk})] = \int_{x_i-h/2}^{x_i+h/2} dx \int_{y_j-h/2}^{y_j+h/2} dy \int_{z_k-h/2}^{z_k+h/2} dz E_a \left( \sum_{l=1}^{N_e} \phi_l(x,y,z)\phi_l^*(x,y,z) \right). \] (15)

Integral (15) can be calculated using the Simpson’s method.

Performing a single time step in the WPMD-DFT or WPMC-DFT algorithm involves three main stages:

(i) calculation of the Coulomb interactions between electrons and ions using equations (4)–(5);
(ii) construction of the spatial mesh and calculation of the energies that depend on the electron density (15);
(iii) solution of the equations of motion or using the Monte-Carlo sampling.

Note that the most essential part of the method is the definition of the interparticle interaction so that the general method name (WPMD-DFT) stems from the MD version of the algorithm, because it implements all aspects of the interaction computation, including energy derivatives with respect to the dynamical variables. However, in most computations reported in this paper we use the Monte Carlo propagation which is less demanding and requires to compute only the energy. We label these computations as WPMC-DFT accordingly.

2.4. Adaptive mesh refinement

The simulation time is determined mainly by stages i and ii that have different dependence on the number of electrons \( N_e \)

\[ \tau(N_e) = \tau_1(N_e) + \tau_2(N_e). \] (16)

When the number of electrons is large (\( N_e > 10^3 \)), the performance is determined mainly by the Coulomb interactions \( \tau_1(N_e) = \Theta(N_e^2) \), so that the simulation time grows quadratically with the number of wave packets. At small electron numbers \( N_e < 10^2 \) the contribution of \( \tau_1 \) becomes negligibly small compared to the time \( \tau_2(N_e) = \Theta(N_e) \) that depends on calculation of the electron density and the numerical integration on the mesh (15).

An increase of the WPMD-DFT method performance can be obtained by a paralleling of the algorithm for both many-core and multi-core computer architectures. The time \( \tau_1 \) may be decreased by using a simple pair decomposition for calculation of the Coulomb forces, while the use of general purpose graphical processing units (GPUs) allows one to substantially accelerate the integration of the exchange-correlation functionals [1]. In addition to this, by representing the integrals in form (14), only local values on the spatial mesh are used, so the spatial domain decomposition becomes straightforward and beneficial for computing clusters with a distributed memory including GPU clusters.

The spatial mesh used to calculate functional (14) covers the volume several times greater than the formal dimensions of the simulation box constrained by reflective potential (6). This is necessary to account for the electron density extending out of the reflective walls, since the reflective potential is soft and the wave packet has nonzero width.

Resolution of the spatial mesh is chosen in a way to optimally balance the performance of the method and the accuracy of computations. In most cases the space step may be defined as
\[ h = s_{\text{min}}/2, \] where \( s_{\text{min}} \) is the minimal wave packet width for the current plasma parameters. The value of \( s_{\text{min}} \) may be estimated form the WP width distribution obtained by auxiliary WPMD simulations without DFT extensions for the same plasma parameters.

For greater densities of plasma, this approach is not applicable, since the original WPMD method leads to non-physical particle configurations (section 3). More universal and more computationally demanding procedure of finding the optimal mesh resolution consists in studying the convergence of the calculated values with the decrease of \( h \).

For the electron number densities \( n_e = 10^{21} - 10^{22} \text{ cm}^{-3} \) and the temperature about \( T \approx 3 \times 10^4 \text{ K} \) we found the optimal space step of \( h \approx 0.1 \text{ Å} \), which corresponds to the spatial mesh of \( M_x M_y M_z \approx 200 \times 200 \times 200 \). These mesh dimensions lead to substantial increase of the integration time \( \tau_2 \) compared to \( \tau_1 \).

The use of a fixed space step is slow and it may lead to computational errors since a wave packet with the width smaller than \( h/2 \) can be located between the mesh nodes do not contribute to the electron density at all. To avoid these errors, it becomes necessary to restrict the packet widths to be large enough when using Monte Carlo method for calculating the thermodynamic properties.

A more efficient alternative to the regular mesh with a constant space step is an adaptive mesh, in which the space step depends on the local electron density. It allows one to reduce the number of computations in the regions of uniform electron density and increase the accuracy in highly non-uniform regions. The most complicated operations for an adaptive mesh construction are mergers and fragmentation of the mesh cells. Generally they require a knowledge of the electron density and its spatial derivatives in the nodes of a high-density regular mesh. However, the use of Gaussian wave packets allows one to estimate the gradient of the electronic density at each space point and perform the adaptive mesh generation without pre-calculating the electron density on a fine mesh.

In our case the adaptive mesh is constructed using the following algorithm. If the simulation region is not cubic, it is split into a set of cubic cells. The cubic cells are subdivided into 8 equal parts (the length of the cell is split into two equal parts in each dimension). The process of subdivision is continued for each subcell until one of the following conditions is met:

- there exists a wave packet at the position \( r_i^\alpha \) in the cell having size \( h \) and center at \( C^\alpha \) (\( \exists i \in 1 \ldots N_\text{e} : C^\alpha - h/2 < r_i^\alpha < C^\alpha + h/2 \), \( \alpha = x, y, z \));
- there exists a wave packet at a distance not greater than \( A s_i \) from its cell border \( C_{\text{edge}} \), where \( s_i \) is the packet width (\( \exists i \in 1 \ldots N_\text{e} : |C_{\text{edge}} - r_i| < A s_i \)).

To avoid an unlimited subdivision of the cells we introduce a condition \( h \geq B \min(s_i) \) which limits the minimal spatial step of the adaptive mesh to the product of the minimal wave packet width in the cell and a parameter \( B \). The minimal width here is found by searching a spheric region of radius \( r = A \min(s_i) \), where \( s_i \) is a width of \( i \)-th wave packet in the cell.

The parameters \( A \) and \( B \) are determined by the form of the wave packet (Gaussian in our case) and do not depend on the physical parameters of the system under study such as density or temperature. These parameters are selected in such a way that the results of numerical integration (14) are not dependent of further varying of \( A \) and \( B \). Initial estimations for \( A \) and \( B \) may be obtained by considering a region where \( \langle \varphi(q) | \varphi(q) \rangle \leq \varepsilon \) for a single wave packet with \( \varepsilon \) being a number close to zero (accuracy parameter). In our work we use the values of \( A = 0.7 \) and \( B = 0.4 \) which provide an optimal trade-off between the accuracy and computational speed.

The described approach allows one to reduce the number of the spatial mesh cells by \( \sim 10^3 \) times for the equilibrium nonideal plasma with the electron density of \( n_e = 10^{21} - 10^{22} \text{ cm}^{-3} \) and temperature \( T \approx 3 \times 10^4 \text{ K} \), which is substantial for the speed-up of the calculations.
2.5. Paralleling the code and LAMMPS integration

As noted above the use of the LAMMPS package is determined by its availability and parallel computation efficiency. The WPMD-DFT algorithm was implemented in a form of LAMMPS pair force field, enabling both original WPMD and WPMD-DFT dynamical calculations for the \( \text{NVE} \) or \( \text{NVT} \) ensembles. Additionally we implemented the Monte Carlo sampling using the \( \text{NVT} \) ensemble.

The use of LAMMPS facilitates developing of the parallel algorithms for WPMD and WPMD-DFT methods. For the original WPMD method, the LAMMPS spatial decomposition of the system volume was utilized by setting the cut-off distance greater than the system size. This setting is formally equivalent to the decomposition of interaction by pairs of particles. Here the term “particles” refers both to the wave packets and classical ions. In this case each computational node has \( N_l \) “local” particles and calculates \( N_l(N_l - 1)/2 \) pair contributions to the potential energy. Additionally, each computational node has information about the locations of \( N_g = N - N_l \) “virtual” particles that are attributed to neighbor cells and calculates the interactions between them and the local ones. Assuming communication overhead being negligible, which is acceptable when the number of particles \( N \) is much greater then the number of nodes, we can estimate the maximal parallel speedup of the original WPMD algorithm for \( p \) computational nodes as

\[
R(p) = p^2(N - 1)/p(2N - 1) - N.
\] (17)

For parallel computations of integral (15) in the WPMD-DFT method we also used the LAMMPS domain decomposition where a computational node is assigned to a corresponding spatial region. Then at each node the adaptive mesh is constructed independently and the partial energy contributions are computed. The decomposition procedure is performed after adjusting the simulation volume for the electron density extending out of the potential walls.

3. Results and discussion

3.1. Influence of the boundary conditions

Although the periodic boundary conditions (PBC) are commonly used in atomistic simulation of homogeneous plasma, in the present work we use a different boundary condition, namely, the confining potential serving as the reflecting boundary. The reason for this is the difficulty in implementation of both periodicity and spatial localization for wave functions such as wave packets. Different types of localization constraints for wave packets in periodic boundary conditions were analyzed in our earlier work [12], and it was shown that all considered constraints either lead to overestimated wave packet widths or required introduction of additional unphysical constraint parameters. The confined system of wave packets that we use in this work is free from these deficiencies and can serve as a simplified model of a quantum system with localized and limited set of basis wave functions.

The use of the WPMD simulation approach with the confinement potential for the study of homogeneous and spatially unconstrained systems requires the procedure of extrapolating the properties of the confined system onto the unconstrained one. For the classical MD plasma simulation with point-like electrons and ions, it has been shown [24] that the effect of the confinement potential compared to the system with PBCs diminishes with the growth of the system size, so that the extrapolation can be performed.

Similarly to the classical MD simulation, the initial density of electrons in WPMD is calibrated as if the electrons were uniformly distributed in the inner part of the simulation box not affected by the confining potential. In this case the electron density is evaluated as a number of electrons divided by volume, where they are located.
Figure 1. Dependence of the electron density profile for a system confined by potential (6) on the number of wave packets in the system for the densities $n_e = 10^{21}$ (a) and $4.22 \times 10^{24} \text{ cm}^{-3}$ (b). Vertical lines are marking the simulation box borders, the horizontal line denotes the mean electron density.

However, the wave packet widths in the WPMD are always under influence of the confining potential. Therefore the real electron density profile in the WPMD simulations (figure 1) differs from the uniform distribution that is usually obtained in the classical MD with PBC. In WPMD the electron density profile is obtained by evaluating expression (10) along the selected axis.

For systems with a small electron density $n_e \lesssim 10^{23} \text{ cm}^{-3}$ the density profile has a bell-formed shape [see figure 1(a)] with a pronounced uniform region in the cell centre and abrupt decrease near the boundaries. Thus the density in the central region is greater than the initial mean density. As the particle number increases, the size of this central region grows and the mean density becomes closer to its initial value. Generally, the larger is the number of particles, the smaller is the density difference and the effect of boundary conditions.

For systems with the density of $n_e \approx 10^{24} \text{ cm}^{-3}$ and larger, the density profile is also bell-shaped [see figure 1(b)], however, with oscillations in the central region. Amplitude of the oscillation decreases with the growth of the particle number, but the peaks near the boundaries still remain.

Convergence of the thermodynamic properties with the system size growth for the WPMD-DFT system in the confining potential (figure 2) is analogous to the one reported in [24]. Besides convergence of the energies, the maximum of the wave packet width distribution also converges with the number of particles (insertion in figure 3). It confirms a self-consistency of the model.

The dependence of the pressure upon the nonideality parameter $\Gamma = (4\pi n_e/3)^{1/3}e^2/(k_B T)$ compared with [16] is shown in figure 4. It is seen that the difference between two methods of pressure evaluation described in section 2.2 is small although equation (8) gives the values of pressure closer to the theoretical curve [26] and the classical MD simulation results [16].

3.2. Internal energy of hydrogen plasmas

The internal energy of the hydrogen plasma was calculated using the original WPMC (Hartree approximation) and WPMC-DFT methods for the temperatures $T = 10^4 – 5 \times 10^4$ K and densities $n_e = 10^{20} – 5 \times 10^{24} \text{ cm}^{-3}$ (figure 5). The Monte-Carlo version of the algorithm was used, since
Figure 2. Dependence of the internal energy of the hydrogen plasma on simulation box size, \( n_e = 10^{21} \text{ cm}^{-3}, T = 3 \times 10^4 \text{ K} \). The dashed line is inverse cubic root fit for \( (E - E_\infty)/N_e \) shifted by \( E_\infty \), where \( E_\infty \) is the internal energy of infinite system.

Figure 3. Distribution of the wave packets by widths; the insertion shows the position of the distribution maximum as a function of the simulation box size, \( n_e = 10^{21} \text{ cm}^{-3}, T = 3 \times 10^4 \text{ K} \).

It is more computationally efficient compared to the dynamical approach and yields the same results assuming the system being ergodic. Compared to the previous work [1] we increased the number of electrons up to \( N_e = 512 \) and thus substantially reduced the sampling error.
Figure 4. Pressure of the hydrogen plasma as a function of the nonideality parameter calculated using equations (8) (red triangles) and (7) (blue triangles). The solid lines represent analytical expressions for the dependence of the pressure on the nonideality parameter: black line is the ideal gas approximation, blue curve is the equation from the work [26]. The green squares are results of classical MD simulations [16]; $P_1 = k_B T (k_B T/e^2)^3$.

Figure 5. Internal energy per particle of hydrogen plasmas as a function of electron density calculated by the WPMC with Hartree approximation and the WPMC-DFT methods.
Figure 6. Execution time for one step of WPMC and WPMC-DFT algorithms as a function of the particle number for a sequential computation (a) and as a function of the number of compute nodes for $N_e = 512$ (b): red circles correspond to the original WPMC method, blue squares and triangles correspond to WPMC-DFT with the regular mesh running on CPU (squares) and on GPU (triangles), aqua squares and triangles correspond to WPMC-DFT with the adaptive mesh running on CPU (squares) and on GPU (triangles). The dashed curves in (a) represent the scalings of $N_e$ and $N_e^2$. The dashed curve in (b) represents the theoretical parallel efficiency (17) for the WPMC algorithm, dot-dashed curve is the ideal parallel efficiency $R(p) = p$.

As one can see in figure 5, at low densities, the results of both methods coincide. Substantial differences are observed at temperatures and densities corresponding to the degeneracy parameter greater than unity. The original WPMC method is unable to correctly reproduce the plasma state at densities above $10^{22}$ cm$^{-3}$ because of non-physical clustering in the electron–ion system which is a computational artefact. This result is conforming to the fact that the density $n_e \geq 10^{22}$ cm$^{-3}$ for all considered temperatures is already beyond the range of applicability for the original WPMC [1]. In contrast to WPMC, for the WPMC-DFT method the exchange-correlation energy increases with the increase of the density and stabilizes the system. Moreover the exchange-correlation contribution leads to a rapid increase of the internal energy at the densities $n_e > 10^{23}$ cm$^{-3}$.

3.3. Performance of the serial and parallel algorithms
Performance of the WPMD-DFT and WPMC-DFT methods while using a regular spatial mesh for calculating the exchange-correlation energy was reported in [1]. In the present work we study the performance for the adaptive mesh and also report on benchmarks for parallel execution of a supercomputing cluster.

Systems with different number of electrons were considered for the fixed electron number density $n_e = 10^{21}$ cm$^{-3}$. For serial execution we used Intel Core i7-7820X CPU and NVIDIA GeForce GTX 1080 Ti GPU. For parallel benchmarks we used the K-100 hybrid supercomputing cluster with Intel Xeon X5670 CPUs and NVIDIA Fermi C2050 GPUs. The results of benchmarks are presented in figure 6.

Additional computations of the exchange-correlation energy slowed down the WPMC-DFT method with the regular mesh by $10^5$ times compared to the original WPMC when using a
single CPU. The use of GPU accelerates the WPMC-DFT computations by $10^2$ times which corresponds to the simulation time of $\sim 10^{-1}$ s per a single time step for the system of 512 electrons.

One can see in figure 6(a) that the mesh computation time $\tau_2$ [in (16)] scales linearly with $N_e$ for $N_e < 1024$. The use of adaptive mesh for the CPU version increases the computational performance substantially and makes it comparable to single GPU computations with the regular mesh. The use of GPU together with the adaptive mesh increases the performance by additional 10 times resulting in $10^{-2}$ s per a single step time. However, it is still $\sim 100$ times slower than the original CPU version of WPMC. The dependence of the performance on the particle number is more complicated for the adaptive mesh, since the actual space size depends both of the widths of the packets and their positions.

Parallel efficiency of the original WPMC method for multi-core CPU systems [see figure 6(b)] is determined mainly by its Coulomb interaction part. For this long-range interaction the interconnection overhead time becomes comparable to the CPU time for 32 compute nodes and 512 electrons.

For the WPMC-DFT method, the dominant computations are related to the spatial mesh, and, therefore the parallel efficiency depends mainly on the mesh algorithm decomposition. At small number of compute nodes the parallel efficiency increases proportionally to the number of nodes. By further increasing the number of compute nodes the parallelism becomes less effective due to the imbalance of the computations between nodes. The highest speedup reached in our calculation is 40 for 128 compute nodes compared to the serial execution.

4. Conclusions
The WPMD-DFT method is the unique simulation approach that handles electron dynamics at high plasma densities. It provides adequate results for an equilibrium hydrogen plasma with the electron number density up to $10^{24}$ cm$^{-3}$. The influence of the confining potential that prevents unbounded wave packet spreading is analyzed. An alternative method for the pressure calculation is proposed.

Development of the adaptive mesh algorithm and using GPU computing allows one to increase the WPMD-DFT performance by 3–4 orders of magnitude but it is still lower than the original WPMD performance. A parallel version of the code is implemented via integration with the LAMMPS package and a reasonable parallel speed-up is achieved. The parallel efficiency is limited mainly by the long-ranged Coulomb interactions.

Acknowledgments
This work is funded by Russian Foundation for Basic Research, project number 19-32-90193 and by the grant in the form of a subsidy for a large scientific project in priority areas of scientific and technological development No. 13.1902.21.0035. Computations were performed at the K-100 cluster (Keldysh Institute of Applied Mathematics RAS, Moscow), Joint Supercomputer Center of RAS and the Supercomputer Centre of JIHT RAS.

References
[1] Lavrinenko Ya S, Morozov I V and Valuev I A 2019 Contrib. Plasma Phys. 59 e201800179
[2] Zaporozhets Yu, Mintsev V, Fortov V, Reinholz H, Röpke G, Rosmej S and Omarbakiyeva Y A 2019 Phys. Rev. E 99 043202
[3] Zaporozhets Yu B, Mintsev V B, Reinholz H and Röpke G 2019 J. Phys.: Conf. Ser. 1147 012099
[4] Sitnikov D S 2019 J. Phys.: Conf. Ser. 1421 012001
[5] Povarnitsyn M E and Levashov P R 2019 Appl. Phys. A 125 688
[6] Rupp P et al 2019 Nat. Commun. 10 1–7
[7] Bystryi R G and Morozov I V 2015 J. Phys. B: At., Mol. Opt. Phys. 48 015401
[8] Raitza T, Röpke G, Reinholz H and Morozov I V 2011 Phys. Rev. E 84 036406
[9] Klakow D, Toepffer C and Reinhard P G 1994 J. Chem. Phys. 101 10766–74
[10] Knaup M, Reinhard P G, Toepffer C and Zwicknagel G 2003 J. Phys. A 36 6165–71
[11] Zwicknagel G and Psch pilgr T 2006 J. Phys. A 39 4359–64
[12] Morozov I V and Valuev I A 2009 J. Phys. A 42 214044
[13] Hansen J P and McDonald I R 1981 Phys. Rev. A 23 2041–59
[14] Morozov I V and Norman G E 2005 J. Exp. Theor. Phys. 100 370–84
[15] Morozov I V, Reinholz H, Röpke G, Wierling A and Zwicknagel G 2005 Phys. Rev. E 71 066408
[16] Bystryi R G, Lavrenchenko Ya S, Lankin A V, Morozov I V, Norman G E and Sa’itov I M 2014 High Temp. 52 475–82
[17] Su J T and Goddard III W A 2007 Phys. Rev. Lett. 99 185003
[18] Jakob B, Reinhard P G, Toepffer C and Zwicknagel G 2007 Phys. Rev. E 76 036406
[19] Jakob B, Reinhard P G, Toepffer C and Zwicknagel G 2009 J. Phys. A 42 214055
[20] Valuev I A and Morozov I V 2015 J. Phys.: Conf. Ser. 653 012153
[21] Morozov I V and Valuev I A 2012 Contrib. Plasma Phys. 52 140–4
[22] Grabowski P E, Markmann A, Morozov I V, Valuev I A, Fichtl C A, Richards D F, Batista V S, Graziani F R and Murillo M S 2013 Phys. Rev. E 87 063104
[23] Plimpton S 1995 J. Comp. Phys. 117 1–19
[24] Lavrenchenko Ya S, Morozov I V and Valuev I A 2016 Contrib. Plasma Phys. 56 448–58
[25] Allen M P and Tildesley D J 2017 Computer Simulation of Liquids (Oxford University Press)
[26] Starostin A N et al (eds) 2004 Encyklopediya Nizkotemperaturnoj Plazmy (Series B vol III-1) (Moscow: FIZMATLIT) pp 247–76