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Localization constraints in Gaussian wave packet molecular dynamics of nonideal plasmas

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Abstract

The problem of wave packet broadening in the method of wave packet molecular dynamics simulations of electron–ion nonideal plasmas is discussed. It is shown that when using a harmonic restrictive potential for the packet widths, simulation results depend strongly on the constraint parameter. Two new approaches to constraining the packet broadening in a less stringent way are analyzed: periodic boundary conditions for widths and a dynamic constraint, based on filtering close particle collisions. These different ways to localize electrons are compared by calculating the dynamical plasma collision rate and the particle pair distribution functions.

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1. Introduction

The method of molecular dynamics (MD) is widely used for simulations of the static and dynamic properties of nonideal (strongly coupled) plasmas in equilibrium and nonequilibrium states. It allows us to study dynamical responses including the dynamical conductivity [1, 2] and the dynamical structure factor [2–5], recombination rates [6], relaxation rates [2, 7–9], etc. Nevertheless, pseudopotentials used for short-ranged interaction between electrons and ions are the subject of controversy. The well-known pairwise models such as Deutsch [10], erf-like [4], corrected Kelbg [11] and other potentials have limited applicability in describing close particle collisions and the effects of electron degeneracy. Most of these potentials imply the approximations needed for mapping the quantum system distribution, described by the Slater sum, onto a system with the classical Hamiltonian. Recently the methods of this mapping have been advanced to statistically reproduce various *ab initio* data, for example the pair distribution functions and the electron degeneracy for the ideal Fermi gas [12]. However, most of the pseudopotentials remain temperature-dependent and contain fitting parameters. This formally restricts MD to the systems close to equilibrium. Moreover,

in spite of the fact that the pseudopotentials work well in the classical plasma limit, their general ability to give insight into the quantum dynamics remains questionable [13].

In contrast to the pseudopotentials having adjustable parameters, the method of wave packet molecular dynamics (WPMD) in its original formulation is a fully *ab initio* method. It was first applied in [14–16] to study the dynamical response of nonideal plasmas. WPMD is thought to capture some of the plasma quantum effects while keeping classical equations of motion in a parametric space and thus providing almost the same computational efficiency as the conventional MD.

The single electron wavefunctions in WPMD are represented by wavepackets (WPs), for which the most computationally efficient is the Gaussian form [16]:

$$\varphi(\mathbf{x}, t) = \left(\frac{3}{2\pi\gamma^2}\right)^{3/4} \exp\left\{-\left(\frac{3}{4\gamma^2} - \frac{\mathrm{i}p_\gamma}{2\hbar\gamma}\right)(\mathbf{x}-\mathbf{r})^2 + \frac{\mathrm{i}}{\hbar}\mathbf{p}\cdot(\mathbf{x}-\mathbf{r})\right\}, \tag{1}$$

where $\mathbf{r}$ and γ determine the WP centre and width, and $\mathbf{p}$ and p_γ are their conjugate momenta respectively. After constructing the full trial wavefunction from the individual one-electron WPs and applying the variational principle to the time-dependent Schroedinger equation, the WP parameters become dynamic variables for the effective classical Hamiltonian. The ions are treated as classical particles. There exist improved WP models with more parameters for the basis wavefunctions [17] or non-Gaussian basis [18].

The most elaborate and computationally demanding way to describe the WP dynamics is to use single determinant antisymmetrization of the total wavefunction for the electrons with the same spin [19]. In the following, discussing the broadening problem (which presumably remains also for the antisymmetrized case), we for simplicity consider the product factorization of the N-particle wavefunction $\Psi(\{\mathbf{x}_k\}, t) = \prod_{k=1}^{N} \varphi(\{\mathbf{x}_k\}, t)$. In this case, the evolution of the WP parameters $\{\mathbf{r}_k(t), \mathbf{p}_k(t), \gamma(t), p_\gamma(t)\}$ follows the solution of the symplectic equations of motion with the Hamiltonian

$$H = \sum_k \left(\frac{\mathbf{p}_k^2}{2m} + \frac{p_{\gamma_k}^2}{2m}\right) + \sum_k \frac{9\hbar^2}{8m\gamma_k^2} + \sum_{k,l} \frac{e^2}{r_{kl}} \operatorname{erf}\left(\frac{r_{kl}}{\sqrt{2\left(\gamma_k^2 + \gamma_l^2\right)/3}}\right), \tag{2}$$

where m is the electron mass and r_{kl} is the relative distance between the particles k and l. From the computational point of view, this method is equivalent to MD with an additional degree of freedom for each electron connected with its variable WP width.

We note that the model construction of the WPMD method is based on the assumption that the total wavefunction (in a product state or an antisymmetrized form) is a combination of contributions from each electron. The essential feature of selecting these contributions in the form of compact WPs is that they are described by a small number of parameters allowing for 'classical' localized interpretation. The drawbacks of this construction are severe limitation of the wavefunction shape and poor description of electrons, delocalized in a phase space. Additional artificial localization constraints, restricting the WP spreading, are often required to keep the WPMD model within its applicability range.

As can be deduced from (2), the WP of a free electron spreads infinitely. The last term in (2) restricts spreading only in the case of strong interaction between an electron and ion, i.e. at high plasma densities and low temperatures. Simulations show that for the plasma parameters considered here ($T \sim 10^4$ K, $n_e \sim 10^{21}\,\mathrm{cm}^{-3}$, $\Gamma = (4\pi n_e/3)^{1/3}e^2/(k_\mathrm{B}T) = 0.5 - 2$), a noticeable number of WPs spread until they completely cease to interact with other particles. It constitutes the main problem of the Gaussian WPMD method and in what follows, we consider different approaches to restrict the width of such WPs.

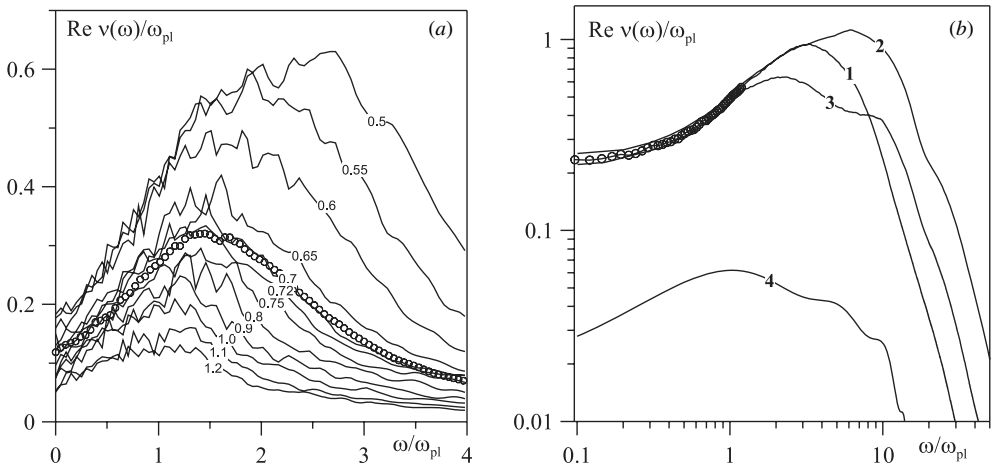

Figure 1. Real part of the dynamical collision rate. (*a*) Fitting of WPMD with the harmonic constraint (solid lines, values of $\gamma_0/\lambda_{\mathrm{th}}$ are shown on the curves) to the MD with the Kelbg potential (circles); $T = 1.35 \times 10^5$ K, $\Gamma = 1$. (*b*) **1** (circles): MD with the Kelbg potential, **2**: WPMD with the harmonic constraint, $\gamma_0/\lambda_{\mathrm{th}} = 0.88$, **3**: WPMD with the energy-based constraint, $U_0/k_{\mathrm{B}}T = 5$, **4**: WPMD with PBS for WP widths; $T = 3 \times 10^4$ K, $\Gamma = 1$.

2. Wavepacket restriction and simulation results

A simple solution proposed in [16] was to introduce an additional harmonic term $\Delta H = \left(9\hbar^2\gamma_k^2\right)\big/\left(8m\gamma_0^4\right)$ to the Hamiltonian (2) which prevents the WP from spreading. It can also be done by changing the imaginary part of the trial wavefunction (1) [20]. The free parameter γ_0 stands for the mean value of γ if there were no Coulomb interaction. In [16] it was taken to be $\gamma_0 = 0.64\lambda_{\mathrm{th}}$, where $\lambda_{\mathrm{th}} = \hbar/\sqrt{mk_{\mathrm{B}}T}$ is the thermal electron wavelength.

As only a rough approximation for γ_0 can be made, it is necessary to check how strong this parameter affects the simulation results. To do that, we consider calculations of the dynamical collision rate $\nu(\omega)$ defined via the dynamic plasma conductivity $\sigma(\omega) = \omega_{\mathrm{pl}}^2\big/[4\pi(\nu(\omega) - \mathrm{i}\omega)]$, where ω_{pl} is the Langmuir frequency. The dynamic conductivity in turn is calculated using the Fourier transformation of the total current autocorrelation function [1, 2] obtained from simulations. The results of WPMD simulations with different values of γ_0 are compared with the results of the conventional MD in figure 1. It is seen that the collision rate depends significantly on the parameter γ_0 which is not strictly defined. The value of $\nu(0)$ for WPMD fits that for MD at $\gamma_0 = 0.72\lambda_{\mathrm{th}}$ for $T = 1.35 \times 10^5$ K and $\gamma_0 = 0.88\lambda_{\mathrm{th}}$ for $T = 3 \times 10^4$ K. The assumption that the relation between γ_0 and λ_{th} is fixed [20] brings us back to the problem of temperature-dependent potentials discussed in section 1.

The additional term in the Hamiltonian which restricts WP widths disturbs the electron trajectories and therefore affects the results for $\nu(\omega)$. To study the broadening we performed test simulations with unrestricted widths, for which periodical boundary conditions (PBCs) were introduced analogous to the conventional PBC for the particle coordinates and with the same period equal to the cell size. It was found that the width distribution converges to a stable limit with an increasing cell size. However, as seen from figures 1(*b*) and 2, this PBC model for the widths leads for Gaussian WPs to a very broad width distribution and severely underestimates interaction.

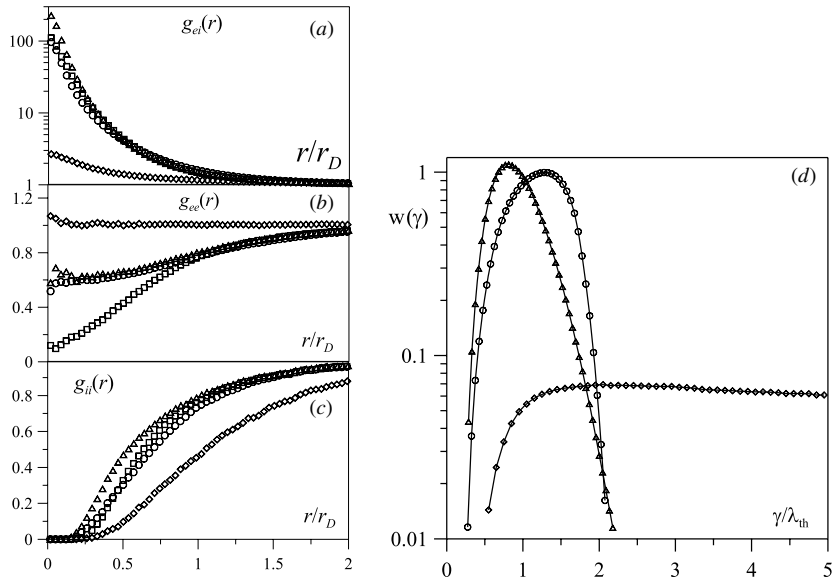

Figure 2. (*a*)–(*c*) Pair distributions and (*d*) the WP width distributions for MD with the Kelbg potential (squares, (*a*)–(*c*) only), WPMD with the harmonic constraint (triangles), WPMD with the energy-based constraint (circles), WPMD with the PBS for WP widths (rhombus). Simulation parameters are the same as in figure 1(*b*).

The idea of the method discussed below is to prevent the WP from spreading, keeping the unperturbed width dynamics during close collisions between particles. This approach follows the general idea of introducing different Hamiltonians for strongly and weakly interacting particles and switching between them during MD simulation [21]. As WP spreading arises only for electrons which weakly interact with other particles, it is reasonable to imply restrictions only for such WPs. The simplest criterion to distinguish between weakly and strongly interacting electrons is to find its binding energy with the nearest ion U_n and compare it with a certain level U_0. A low value of $U_n \leqslant U_0$ means that the electron either undergoes collision or it is temporarily trapped by the ion. All electrons with $U_n > U_0$ are marked as 'free' ones.

The technique proposed here is to add a reflecting wall to the Hamiltonian (2) $\Delta H = A(\gamma - \gamma(t_0))^6 \Theta(\gamma - \gamma(t_0))$ when an electron moves away from the strong interaction area ($U_n > U_0$), where t_0 is the time moment when we mark this electron as 'free', A is a dimensional coefficient and Θ is the Heaviside step function. If the interaction becomes stronger ($U_n \leqslant U_0$), the restriction is switched off. It is possible to select U_0 so that $\nu(\omega = 0)$ matches the result for the Kelbg potential (figure 1(*b*)). As seen from figures 2(*a*)–(*c*), the type of WP restriction strongly affects the pair correlation function. In particular, the values of $g_{ei}(0)$ are close for the Kelbg potential and the proposed WP restriction whereas it is higher for the harmonic constraint. This indicates that the WP constraint must be selected in a more rigorous way and probably with the reference to the many-particle *ab initio* data.

Antisymmetrized construction of the total wavefunction accounts for exchange effects. In this case, the resulting dynamical variables cannot be directly attributed to the individual electrons and the electrons are in some sense delocalized by construction. The AWPMD simulations of the hydrogen plasma [19] have shown that exchange effects may work as a restricting factor for the WP widths. No additional localization constraints are found to be

necessary for AWPMD at least for the temperature and density range studied in [19] ($T \leqslant$ 5000 K). Our preliminary results however show that at higher temperatures (above 3×10^4 K), the problem of WP broadening remains even for the antisymmetrized model.

3. Conclusions

The problem of wave packet spreading in the WPMD simulations of electron–ion nonideal plasmas is analysed. It is shown that in the case of non-antisymmetrized simulations, the simple WP width restriction using the additional harmonic term in the Hamiltonian leads to high sensitivity of the results for the dynamical collision rate (dynamical plasma conductivity) to the model parameter γ_0. Introducing periodical boundary conditions for the WP widths may be thought of as a parameter free method to restrict the spreading of weakly coupled WPs. However, this method leads to unsatisfactory collision rates and pair correlation. We also discussed a restriction technique based on the electron–ion interaction energy, which does not disturb the electron trajectories during close collisions.

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References

[1] Morozov I, Reinholz H, Roepke G, Wierling A and Zwicknagel G 2005 *Phys. Rev.* E **71** 066408
[2] Morozov I V and Norman G E 2005 *J. Theor. Exp. Phys.* **100** 370
[3] Hansen J P and McDonald I R 1981 *Phys. Rev.* A **23** 2041
[4] Zwicknagel G and Pschiwul T 2003 *J. Phys. A: Math. Gen.* **39** 4359
[5] Golubnychiy V, Bonitz M, Kremp D and Schalges M 2001 *Phys. Rev.* A **64** 016409
[6] Lankin A V and Norman G E 2009 Fluctuation approach in the theory of strongly coupled plasmas *J. Phys. A: Math. Theor.* **42** 214042
[7] Zwicknagel G 1999 *Contrib. Plasma Phys.* **39** 155
[8] Morozov I V and Norman G E 2003 *J. Phys. A: Math. Gen.* **36** 6005
[9] Murillo M S and Dharma-wardana M W C 2008 *Phys. Rev. Lett.* **100** 205005
[10] Deutsch C 1977 *Phys. Lett.* A **60** 317
[11] Filinov A V, Bonitz M and Ebeling W 2003 *J. Phys. A: Math. Gen.* **36** 5957
[12] Dharma-wardana M W C and Murillo M S 2008 *Phys. Rev.* E **77** 026401
[13] Jones C S and Murillo M S 2007 *High Energy Density Phys.* **3** 379
[14] Klakow D, Toepffer C and Reinhard P-G 1994 *J. Chem. Phys.* **101** 10766
[15] Knaup M, Reinhard P-G, Toepffer C and Zwicknagel G 2003 *J. Phys. A: Math. Gen.* **36** 6165
[16] Zwicknagel G and Pschiwul T 2006 *J. Phys. A: Math. Gen.* **39** 4359
[17] Lenglet A and Maynard G 2007 *Nucl. Instrum. Methods Phys. Res.* A **577** 343
[18] Murillo M S and Timmermans E 2003 *Contrib. Plasma Phys.* **43** 333
[19] Jakob B, Reinhard P-G, Toepffer C and Zwicknagel G 2007 *Phys. Rev.* E **76** 036406
[20] Ebeling W, Filinov A, Bonitz M, Filinov V and Pohl T 2006 *J. Phys. A: Math. Gen.* **39** 4309
[21] Valuev I 2006 *J. Phys. A: Math. Gen.* **39** 4465