Molecular dynamics with atomic transitions and nuclear reactions

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Abstract. We describe molecular dynamics particle simulations with particles that have internal structure and/or undergo reactions. These calculations give an atomic-scale description of hot plasma based on well-established microphysics and test kinetic theories used to calculate energy exchange and transport in plasma hydrodynamic simulations. The computer experiments can be given detailed diagnostics, without the usual limits of resolution of Laboratory equipment. Typical applications are non-equilibrium atomic kinetics for emission or absorption of X-ray laser radiation by solid targets and/or atomic-scale simulation of hot plasma with fusion reactions, as in inertial fusion ignition experiments.

1. Introduction

Molecular Dynamics (MD) particle simulation using large digital computers was introduced by Alder and Wainwright in 1957 [1]. MD studies a system of particles which represent atoms, molecules or ions. Each particle has classical coordinates \( R_i(t), V_i(t) \), and moves according to Newton's laws governed by forces from the neighbor atoms. For many years the particles were structureless hard spheres or point charges and the goal was to study particle correlations and phase transitions. Early calculations showed that computer particles rapidly relax to an equilibrium Maxwell distribution as a result of binary collisions [2]. In 1966, Brush, Sahlin and Teller applied the complementary technique of Monte Carlo (MC) simulation for the equilibrium statistical mechanics of point charges in dense plasmas [3]. The MC method is also widely used for dynamical processes.

Here we consider particle simulation in which the particles can change state. For example, if the particles are atoms or ions with excited states, we can study atomic kinetics and emission and absorption of radiation. If the particles are low-Z hydrogen isotopes, we can study fusion reactions and fusion ignition [4]. The particle motion in our simulation is classical Molecular Dynamics while the quantum transitions are treated by a Monte Carlo method (see Appendix A, Appendix B). The composite simulation can give new insights for a wide class of important scientific questions.

A fundamental scientific challenge of the calculations is the interface between classical and quantum descriptions: how can we include quantum effects, as completely and accurately as possible, in a basically classical particle simulation? Can we treat the particle motion by classical mechanics, turn the calculation over to quantum mechanics for an atomic transition or reaction, and then revert to classical dynamics for the reaction outputs? What is the best way to handle these abrupt changes of description (see Appendix B)?
A second fundamental challenge concerns computational efficiency. What new computational methods are needed to simulate atomic-scale many-body systems with reactions? We need to add or subtract particles created or destroyed in nuclear reactions; we want to use smaller time-steps for fast-moving parts of the system, and would like to use importance-weighted sampling for efficient computation (see Appendix A). A strategy is needed to handle many atoms when each atom can make many possible transitions between many excited states.

Fusion simulation must include ion-ion collisions, electron-ion and electron-radiation coupling, fusion alpha energy-loss and the production of energetic "knock-on" recoil ions, and the fusion reactions should be calculated for the actual ion velocity-distribution when this is not a simple Maxwellian. In section 3 we describe calculations of fusion ignition at inertial fusion target conditions and illustrate some scientific conclusions we can draw from these calculations.

2. MD code structure

The codes described here have the usual structure of Molecular Dynamics particle simulations [5]. They treat a small volume inside an assumed infinite medium, imposing periodic boundary conditions that keep the particles inside the simulation box. Sub-boxes are used to efficiently locate other atoms near any given atom. For our calculations, the simulation box is divided into 3375 = (15)^3 sub-boxes. Atoms move freely between the sub-boxes.

Pointers (ion --> box, box --> neighbor boxes, box --> ions) serve to locate the atoms. For each atom one pointer tells which sub-box it is in, a second pointer identifies the 26 neighbor sub-boxes, and a third pointer tells which ion is in each sub-box. The pointers are updated during each time-step. A simple calculation gives the periodic distance between two particles, i.e., the distance calculated the shortest way in the toroidal topology of the periodic boundary conditions. Neighbor boxes are defined consistent with periodic boundary conditions.

Fast-moving particles take smaller time-steps than normal particles and those subcycle time steps are synchronized to the main time-steps. For example, in fusion calculations each fusion alpha particle should take 10 time steps during one time-step for the majority D,T ions because its velocity is ~ 10 times larger. The motion of slow particles is interpolated to supply position data for the subcycle time-steps.

In the usual kinetic theory of gases, the internal state of atoms is assumed to be separate from or uncoupled to the motion in the gas. The MD simulation is able to study the effects of coupling between particle position, velocity and internal state.

The internal state of atoms or ions is coupled to position and velocity \( \mathbf{R}_i, \mathbf{V}_i \) in various ways: A fast-moving atom has more chances for collision than a slow-moving atom, and the cross-sections for reactions depend on the relative velocities \( | \mathbf{V}_1 - \mathbf{V}_2 | \) of the reacting particles. The Doppler shift of emission lines depends on the atom's velocity. In a plasma or ionized gas, the electric field \( \mathbf{E}(\mathbf{r}) \) of nearby ions causes a Stark-effect splitting of spectral lines and the Stark profile depends on the atomic position \( \mathbf{R}_i \). Laser gain profiles and escape factors depend on the statistical distribution of velocities and line-widths in the whole plasma. There are simple theories for most of these couplings, but the theories typically treat one process at a time and/or assume a Maxwell distribution of velocities. Molecular Dynamics can relax these assumptions and test the theories.

3. Atomic kinetics for MD

We compare three methods to compute the atomic kinetics. These methods aim to calculate the statistical distribution of excited states, which is summarized by

\[
N_j = \text{probability that atom } i \text{ is in state } j \quad (j = 0, 1, 2, \ldots)
\]  

Method 1.) Schroedinger Equation.

It is possible to solve a numerical Schroedinger equation for each atom i:
Here $H_i$ is a one-atom Hamiltonian function. $U_i(t)$ is time-evolution operator which tells how the wave-function $\Psi_i(t)$ evolves. Projected onto $\psi_j$, which is the eigenstate for the excited state $j$, this gives the population

$$N_{i,j}(t) = \left| \langle \psi_j \left| U_i(t) \right| \Psi_i(0) \rangle \right|^2$$

In recent years the numerical Schroedinger eqn. has been used to study harmonic generation by high-intensity laser radiation. Reference [6] describes a one-atom Schroedinger code used to study mixed Stark and Zeeman effects for a hydrogen atom subject to high-frequency microwave radiation. For electron cyclotron resonance (ECR) heating in magnetic fusion plasma experiments, the microwave frequency is nearly resonant with the atomic Zeeman splitting.

The Schroedinger equation describes time-resolved atomic kinetics, i.e., it describes the time evolution of radiation emission or electron impact excitation. It would be possible to modify the Schroedinger equation to describe rapid transitions that are not time-resolved by introducing an effective Hamiltonian operator.

Using the Schroedinger equation, it is possible to study certain phenomena not encompassed by the other methods, for example: time-dependent Stark splitting [6] in the local atomic environment, or ion-ion collisions during emission of light and the consequent modification of Doppler line-width [7].

Method 2.) CR Method.

It is also possible to solve many copies of the Collisional-Radiative (CR) atomic kinetics model

$$\frac{dN_{ij}}{dt} = -N_{ij} \sum_k T_{j\rightarrow k} + \sum_k N_{ik} T_{k\rightarrow j}$$

The transition rate-matrix is schematically indicated by

$$T_{j\rightarrow k} = n_e \langle \sigma_{jk} v_e \rangle + A_{jk} (n_{hv} + 1)$$

where $\sigma_{jk}$ is the electron collision cross-section to change the atomic state $j \rightarrow k$, $A_{jk}$ is a radiative rate (written for emission), and $n_{hv}$ is the number of photons per mode in the radiation environment. The notation is schematic because there are many additional processes [8]:

- **Electron collisional excitation**
- **Collisional de-excitation**
- **Electron-impact ionization**
- **3-Body recombination**
- **Absorption of radiation (excitation)**
- **Emission**
- **Photo-ionization**
- **Radiative recombination**
- **Autoionization**
- **Dielectronic recombination**
- **Stark ionization**
- **Stark recombination**
For several of these processes, simulations with classical point electrons have difficulty to obey detailed balance, and this is an argument in favor of the fluid description of free electrons which we adopt in this work.

The one-atom Collisional-Radiative model is a well-established computational method and has been used to analyze spectra from astrophysics and magnetic fusion plasmas for almost 50 years with extensive comparison to experiments [8]. The applications have required detailed knowledge of atomic processes, and useful collections of atomic data are available to accurately describe specific atoms and ions [9].

The CR rate equation describes atomic processes as instantaneous transitions; this is the opposite extreme from the Schrödinger equation. We should distinguish the rate of transitions (which depends on density) from the duration of the transition event. The actual duration of transitions depends on the atom (ion) considered, but roughly speaking,

\[\begin{align*}
e-i \text{ collision} & \sim 10^{-17} \text{ sec} \\
i-i \text{ collision} & \sim 10^{-15} \text{ sec} \\
radiation & \sim 10^{-12} \text{ sec}
\end{align*}\]

We underline that the transition duration depends on the ion and the temperature; for example, radiation emission takes as long as 10 nsec for hydrogen (Z = 1) but is much quicker for high-Z ions, while "forbidden" transitions have lifetimes as long as 1 sec and are only seen from atoms which are not often disturbed by collisions in low-density astrophysical plasmas.

Method 3.) "Markov" Method.

In this method, each atom i is assumed to be in a definite excited state \(j(i)\) at each time. The state can change during each time-step, governed by probabilities; this is a mixed MC/MD method. The probability of the transition \(j \rightarrow k\) during \(dt\) is taken to be

\[P_{jk} = T_{j \rightarrow k} dt\]  \hspace{1cm} (6)

The time-step \(dt\) must be small enough so that the sum (over \(k\)) of the probabilities is less than unity for all initial states \(j\). In fact, for most excited states, on most time-steps, there is no transition; for efficiency, the MC test considers this most-likely case first.

Since this method appears to assume the atomic state is constantly being measured, there's a natural question whether it gives the same answer as the other methods. In general, we expect the Markov method to be entirely equivalent to the CR equations but to neglect quantum interference effects that are included in the Schrödinger method. An example of such a quantum effect is the mixing of 2s and 2p states that occurs when an atom is in a local electric field (Stark effect) [6]; this mixing effect has no representation in the CR or Markov descriptions.

3.1 Test of the Markov method

We describe a simple test of the Markov method for atomic kinetics by comparison with the usual CR model.

We consider neutral hydrogen, and describe the atomic dynamics by the populations of the 1,012 excited states having principal quantum number \(n = 1\) to \(n = 11\). The CR calculation follows the evolution of one atom, and finds the probabilities for the excited states. The atom begins in the \(n = 1\) groundstate. The transition rates obey detailed balance consistent with a plasma temperature of \(T_e = T_i = 10\) eV. The excited state populations relax to thermal equilibrium (with the expected Boltzmann distribution) in one nsec. (This is not complete equilibration because there is no coupling between electronic excited states and the ion kinetic energy, so the ion temperature remains zero in this test.)
We repeat the same calculation by the Markov method, using the same set of excited states and the same rates $T_{jk}$. It was possible to perform the calculation on a desktop computer for a plasma containing 100,000 atoms. For this test, all the atoms are identical so the result should be the same as the CR calculation. The agreement is very good (Figure 1). If the atoms were moving or in different environments, the MD calculation could include those effects and presumably would be more accurate.

The most interesting result is the observation that, using the Markov method, the MD+CR calculation is feasible.

### 3.2 Practical limits

First, treating electrons as classical particles would require a very small time-step $dt \sim 10^{-21}$ sec, because without this time-resolution the calculation of the strongly curved orbit of electron scattering by a nucleus will not be accurate enough to conserve energy. Because such short time-steps severely limit the calculation, we adopt a fluid description of free electrons, described below as a **Langevin method**.

The Schrödinger equation description also requires a small time step,

$$dt << \frac{\hbar}{|E_j|k} \sim (5 \times 10^{-17} \text{ sec})/Z^2$$

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**Figure 1.** Comparison of one-atom collisional-radiative (CR) model for hydrogen with the Markov MD model described in the text. For this comparison the atoms are not moving and all have the same local environment. The 1012 states used and transition rates are the same in the two calculations; the purpose is to test the Markov MD method to calculate atomic kinetics. Agreement with the CR method is excellent, but similar calculations with smaller numbers of atoms showed larger effect of fluctuations, especially for excited states with small populations. The most interesting conclusion is that even 1000 excited states and 100,000 atoms represent a feasible calculation on a desktop workstation.
where $E_j$ is the binding energy of an atomic state. With such a small time-step, the plasma simulation can only be followed for relatively short times. The minimum storage required for the Schrödinger eqn. calculation is something like

$$\sim (6N_{\text{atoms}}) \times (2N_{\text{states}})$$

i.e., six position-velocity variables for each atom and the real and imaginary parts of the probability amplitude for each excited state. In fact, most MD codes have several copies ("old" and "new") for each of the variables, so the storage is proportional to the number indicated above.

For the collisional-radiative model, the time-step must at least resolve the transition rates,

$$dt \ll 1 / T_{jk} \sim 10^{-12} \text{ sec}$$

The time-step could be further restricted by the MD atomic motion, depending on the density; ions should not be able to cross a sub-box during one time-step to avoid mis-counting their collisions. The storage needed is proportional to $\sim (6N_{\text{atoms}}) \times (N_{\text{states}})$ an estimate which includes atomic position-velocity variables and the probability $N_{ij}$ for the excited states on each atom.

In contrast, the Markov method stores only one integer for each atom, as if each atom is instantaneously "in" one state. The time step $dt \ll 1 / T_{jk} \sim $ psec is similar to the CR model but the required storage is much less:

$$\text{Storage } \sim 7 N_{\text{atoms}}$$

However with this method one needs to average over many atoms to reduce the fluctuations, which are especially visible for excited states with small populations.

### 3.3 Langevin Description of free electrons

For practical reasons mentioned above, we seek to replace particle electrons by an effective fluid which will represent the behavior of the electron gas over longer time-scales. The fluctuations in this fluid produce important electron-ion energy-transfer effects, so we must keep track of the fluctuations. For the present we assume the electrons have equilibrium Maxwellian distribution at the current electron temperature $T_e(t)$, and work out the fluctuations from that assumption, using statistical methods described by Chandrasekhar [10].

Electrons equilibrate much more rapidly than ions, so it is possible to assume Maxwellian electrons even while allowing non-Maxwellian ions.

We describe the Langevin treatment of ion coupling to electrons. During one ion time-step $dt$ there are many electron-ion collisions. The momentum transfers from these collisions combine as a random walk to produce a total momentum change $d\mathbf{p}_{\text{tot}} = M_i \mathbf{v}$ during time $dt$, the probability that the ion changes velocity by $d\mathbf{v}$ as a result of the electron-ion collisions is

$$P(\mathbf{v}, d\mathbf{v}, dt) = \left(\frac{M_i \tau}{2\pi kT_e dt}\right)^{3/2} e^{-\frac{M_i \tau}{2kT_e dt}\left(d\mathbf{v} \cdot \mathbf{v}\right)^2}$$

(7)

Here the parameter $\gamma = dt / 2 \tau$ where $dt$ is the time-step and $\tau$ is the collision time given below. The average (over this probability distribution) of the change of ion velocity is

$$< d\mathbf{v} > = -\gamma \mathbf{v} = -\left(\mathbf{v} / 2\tau\right) dt$$

(8)
Eq. (8) describes the ion energy-loss $dE/dx$ to electrons. If the electrons are hotter than the ions, the random transfers $d\nu$ will heat the ions. Eqn. (7) has been written for the case in which the ion velocity is not large compared to the electron thermal velocity. The electron-ion collision time $\tau$ that results from the statistical calculation is:

$$\frac{1}{\tau} = \frac{8\sqrt{2}\pi}{3} \frac{Z^2e^4n_e}{MkT_e} \sqrt{\frac{m_e}{m_i}} \log \Lambda$$

(9)

This well-known collision time goes under the names of Landau and Spitzer.

The Langevin method gives a real benefit for the time-step in an MD code; in our fusion ignition simulations of multi-keV plasmas we can use a time step of $10^{-17}$ sec which makes it possible to carry a calculation for as long as 10-20 psec.

The time-step is a concern even for the largest parallel supercomputers, because while the supercomputer can do many calculations at the same time, the largest number of sequential time-steps that can be chained together (on today's machines) is only of the order of $10^6$ or $10^7$.

3.4 Applications of the MD + CR method

Most of the applications will be studies of dynamical or non-equilibrium phenomena. We can offer a list of likely prospects:

- X-ray laser interaction with cold matter [11]
- Gas laser gain profiles; gain narrowing
- Laser cooling; coherent emission by neighbor atoms (super-radiance)
- Tokamak edge plasmas; collisional modification of Doppler profiles
- Astrophysical plasmas, interstellar medium
- Combustion Science ($H_2$, $O_2$ combustion)
- Fusion ignition and novel fusion schemes

Of these possibilities, only the fusion ignition particle simulation has been worked out in detail.

4. Fusion Ignition Simulations

Fusion ignition is a suitable application for molecular dynamics, because the atomic-scale physics of fusion involves a small number of essential species: deuterium and tritium ions, electrons, radiation and fusion reaction product alphas and neutrons [12, 13, 14].

We describe fusion simulations performed with a hybrid MD/MC computer code named OK (for Okehazama, a historic spot in Aichi prefecture). Table I gives a brief overview of the physical processes included in the code.

Ten thousand D,T ions are described by their positions and velocities \{\textbf{R}(t), \textbf{V}(t)\}. At keV temperatures the DT is fully ionized and there are no impurities other than fusion output alpha particles. The velocity distribution is constrained only by collision processes in the code. An "ion temperature" $T_i(t)$ is calculated as 2/3 of the instantaneous average ion kinetic energy.

Special computational techniques used in the OK code include:

1.) Fast-moving $\alpha$ particles are subcycled (synchronized small time-steps)
2.) Electrons and ions are coupled by the Langevin method described above.
3.) Electrons emit and absorb bremsstrahlung radiation, calculated with Kramers' cross-sections [15]
4.) DT ion pair-collisions are treated by an analytic method
5.) Fusion reactions occur based on a Monte Carlo test imposed during each DT collision.
6.) The Monte Carlo tests are "guided" to reduce fluctuations (see Appendix A).
The plasma composition changes with nuclear reactions. A flag or pointer vector identifies whether each ion is currently "active" or "consumed". When DT ions react they are promptly rendered inactive. The ion list is not rearranged, but rather the "active ion" flag is switched and that ion is skipped for all further processes and audits. Reacting D and T ions are replaced by a fusion alpha and neutron having high kinetic energies.

The Monte Carlo method to test for fusion is described in Appendix B.

4.1 Ignition simulations

We describe simulations of inertial fusion ignition using assumed inertial fusion target hot-spot parameters,

$$DT \text{ fuel density} = 100 \text{ g/cm}^3 = 2.41 \times 10^{25} \text{ ions/cm}^3$$

Temperatures begin near 10 keV (electrons and ions) and 1 keV (radiation)

Laboratory experiments performed to date do not quite achieve these desired initial conditions [16, 17].

It is useful to describe the physical conditions in such a hot dense plasma in order to provide an intuitive idea about which physical processes are most important.

At density of 100 g/cm$^3$, there are $\sim 2.4 \times 10^{25}$ ions/cm$^3$. The Debye length is $\sim 0.1$ nm and the (usual) Coulomb logarithm $\sim 4.5$. At the plasma ion density $N_i$ considered here, the ion-sphere radius ($\sim$ half the distance between neighbor ions) is $R_o = (3/4\pi N_i)^{1/3} \sim 0.02$ nm. For $kT \geq 10$ keV, the ion coupling parameter

$$\Gamma = Z^2 e^2 / R_o kT \leq 0.007$$

Table 1. Physical processes included in the OK code simulations described in the text.

| OK Code PHYSICS |
|-----------------|
| **IONS**: $N_i = 10,000$ DT IONS | Described by classical $\{ R_i(t) \}$, $\{ V_i(t) \}$ |
| **ELECTRONS**: $N_e = N_i$, $T_e(t)$ | The electron velocity distribution is assumed to be instantaneously Maxwellian |
| **RADIATION**: $\{ I_\nu \}$ | Isotropic, homogeneous radiation is described by a 200 group photon frequency spectrum. Radiation is emitted and absorbed during electron-ion collisions; Kramers' bremsstrahlung cross-sections are used. |
| **ALPHAS**: Variable number $N_{\alpha}(t)$ | $\{ R_\alpha(t) \}$, $\{ V_\alpha(t) \}$ Move in plasma with subcycle time-steps $dt = dt_{ef} / 10$. |
| **NEUTRONS**: Collected in output spectrum $P(v)$ |

**Processes:**

- Ion-ion Collisions
- DT fusion reactions
- Electron-ion collisions
- Emission, absorption of radiation
Figure 2. Energy changes during the ignition calculation reported as Case A in the text. The total energies of 10,000 DT ions (and electrons, radiation and fusion alphas) are reported in electron volts (eV). Careful inspection shows that while the alpha energy increases after each fusion reaction, it decreases during the intervals between fusions due to energy deposition in the plasma.

This inequality shows that equilibrium ion pair-correlations due to the Coulomb potential, which would be important at lower temperature, do not have a large effect on plasma at fusion ignition conditions. The Fermi energy ~ 300 eV << kT so the plasma is entirely non-degenerate. Strong ion-coupling and degeneracy effects are more important during assembly (implosion) of the fuel.

The computational box size is L = 0.75 nanometers. With DT ion velocities $v_D \sim v_T \sim 10^8$ cm/sec, the time taken to cross a sub-box (L/15) is about 5 $10^{-17}$ sec, i.e., five time-steps.

Ion distribution becomes approximately Maxwellian within ~ .1 - .2 psec due to ion-ion collisions, but the high-energy tail of the distribution may not be Maxwellian.

Fusion alpha energy-deposition requires ~ 2 - 5 psec. Because of this delay, fusion ignition requires ~ 10-20 psec.

We describe results from 4 ignition self-heating calculations which start from slightly different initial temperatures and run for different times. (Figure 2) All these calculations assume a constant DT fuel density 100 g/cm$^3$. In each case the radiation begins as a 1 keV blackbody and heats to ~ 5 keV but is not a Planck spectrum.

| Case | Initial temperatures ($T_e^0 = 8$ keV, $T_i^0 = 9$ keV) | 123 fusions during 15 psec | clock time = 84 h | Final temperatures ($T_e^f = 18.7$ keV, $T_i^f = 16.6$ keB) |
|------|----------------------------------------------------------|---------------------------|-------------------|-------------------------------------------------|
| A    |                                                          |                           |                   |                                                 |
| B    | Initial temperatures ($T_e^0 = 9$ keV, $T_i^0 = 11$ keV) | 100 fusions during 10 psec | clock time = 60 h | Final temperatures ($T_e^f = 17.3$ keV, $T_i^f = 15.7$ keV) |

| Case | Initial temperatures ($T_e^0 = 8$ keV, $T_i^0 = 9$ keV) | 123 fusions during 15 psec | clock time = 84 h | Final temperatures ($T_e^f = 18.7$ keV, $T_i^f = 16.6$ keB) |
case C  Initial temperatures  \( T_{e0} = 10 \text{ keV}, T_{i0} = 7 \text{ keV} \)
45  fusions during 10 psec  clock time = 48 h
Final temperatures  \( T_{ef} = 12.5 \text{ keV}, T_{if} = 10.4 \text{ keV} \)

case D  Initial temperatures  \( T_{e0} = 11 \text{ keV}, T_{i0} = 9 \text{ keV} \)
316  fusions during 20 psec  clock time = 89 h
Final temperatures  \( T_{ef} = 32.2 \text{ keV}, T_{if} = 31.9 \text{ keV} \)

These calculations were repeated with different random-number seeds using the fluctuation-suppression algorithm described in Appendix A. The results were the same to 1 or 2 %.

4.2 Fusion "ignition attractor" phenomenon
In comparing several ignition calculations (like cases A-D) with similar starting conditions, it is found that the plasma rapidly relaxes to a definite ignition heating trajectory and self-heats along this well-defined path in the space of \( T_e, T_i, T_r \). The heating path can be considered to be an attractor in the parameter space [4].

4.3 Non-Maxwell ion distribution
Most fusion events involve an ion that has energy \( \gg kT \). While the bulk of the ion distribution is quite close to a Maxwellian (at a changing temperature), the most important ions are a small minority in the high-energy tail of the distribution, which we consider to be the range from 2 \( kT_i \) to perhaps 10 \( kT_i \). Molecular dynamics is able to calculate this minority population with no \textit{a priori} assumption aside from the geometry and boundary conditions.

A non-Maxwell ion distribution \( f(E) \) can result from
1.)  \textit{Knock-on} DT ions at 20-100 keV produced by Coulomb collisions with fast alpha particles. As the fuel temperature rises and energy loss \( dE/dx \) to electrons decreases, this process becomes more important.
2.)  Ion beam heating (fast-ion fast-ignition). Comparisons have found that the knock-on DT ions assist simple plasma heating to reach a self-sustaining reaction.
3.)  One might expect that if the plasma temperature would rise too fast, there would not be enough time for tail-filling, leading to a depleted tail. In fact the MD calculations do not seem to support this expectation, probably because the heating rate is itself limited by the same couplings that relax the ion velocity distribution.
4.)  In real fusion targets, the heated material has a finite \( \rho R \). In that case, spatial loss of high-energy DT ions depletes the active tail of the DT distribution and can significantly reduce the fusion yield. This mechanism was not included in the calculations described here but can readily be included in future work. (See Petschek & Henderson [18], Nishikawa, Takabe & Mima [19], and Molvig, Hoffman et al. [20])

4.4 Neutron TOF signal
The clearest experimental signature of non-Maxwellian ion distributions would be in output neutron velocity spectra (time of flight spectra). The MD calculations described here predict a small pulse of energetic neutrons that move faster than the tail of a Maxwellian curve-fit [4].

The MD predicts one in \( 10^6 \) of the neutrons will have velocity
\[
\delta v \sim 5 \times 10^7 \text{ cm/sec}
\]

faster than the neutrons in the tail of a Gaussian curve-fit to the overall neutron velocity spectrum. This is only a small fraction of the neutrons but may be easy to detect because they arrive early. At a distance 10 meter away from the fusion capsule, these fast neutrons would arrive \( \sim 2 \) nsec early.
4.5 Code tests

As the OK code was constructed, the physical processes were tested one by one to verify that they give reasonable rates and properly obey detailed balance. The results of these tests are briefly summarized:

I. Ion-ion collisions relax an initial non-Maxwellian ion distribution to a Maxwellian in about the expected time. This comparison is an important test of the small-ball method for ion-ion collisions. For the test, a delta-function distribution \( f(E) \) is evolved by a calculation which includes only ion-ion collisions. The relaxation is examined two ways: the overall ion velocity distribution (plotted as a histogram) is compared to the expected Maxwellian and three moments of the distribution are calculated and seen to approach the expected Maxwellian limits. The time to evolve from a delta-function distribution to a good Maxwellian is about 0.12 psec when the ion temperature is 5 keV (density = 100 g/cm\(^3\)), and ~ .35 psec when the temperature is 10 keV.

II. The electron-ion (Langevin) coupling is tested by a special calculation in which the electron temperature is held fixed, ion-ion collisions are suppressed, and the only coupling is the Langevin coupling between electrons and ions. In this calculation the ion distribution relaxes to a Maxwellian at the electron temperature. This relaxation takes as long as 5 psec for \( T_e = 5 \) keV, significantly slower than the ion-ion thermalization.

III. The radiation emission/absorption formulas used here obey detailed balance and would eventually relax the photon distribution to an equilibrium Planck distribution [15]. Radiation equilibration is much slower than ion-ion or electron-ion equilibration and radiation is far from equilibrium throughout the ignition calculations.

IV. Energy conservation is an important test. The code conserves energy to a few % even on long calculations, approximately one part in \( 10^8 \) per time-step.

4.6 Limitations

The OK code has several limitations: The ion-ion collision algorithm assumes weak ion coupling and uniform electrons. This assumption is appropriate for fusion ignition conditions, where \( \Gamma \ll 1 \), but would not be justified at lower temperature (implosion conditions). For this reason the code is suitable above \( T_{e,i} = 2 \) or 3 keV, but would need modification for lower temperatures.

There are statistical fluctuations \( \sim 1/N^{1/2} \). With \( 10^4 \) ions, the fluctuations in the main plasma properties are not too large. For example, when we plot separate temperatures for D and T ions, these show fluctuations \( \sim 1-2 \% \). However there are only about 100 fusions in a typical calculation and this number shows fluctuations \( \sim 10 \% \), as verified by repeating runs with various random-number seeds. A technique to reduce these fluctuations is described in Appendix A.

4.7 Verification of hydrodynamic simulation codes

Large-scale inertial fusion experiments have been designed and are modeled by elaborate "multiphysics" hydrodynamic simulation codes. These codes represent a remarkable investment of scientific expertise and have been tested over many years on a large variety of laser (and other) experiments [21].

However, fusion experiments performed on the NIF laser do not (today) agree with the predictions of "multiphysics" hydrodynamic simulations [16,17]. There could be several reasons for the disagreements, some involving the calculations, others involving the laser itself or the target fabrication.

It should be clear that the independent calculation of ignition in simple homogeneous fuel by the MD simulation does not directly give much information about target experiments, but could be useful to help identify aspects of the hydrodynamic simulations that need improvement.
5. X-Ray Laser Interaction with cold matter

H. Yoneda has recently described experiments on the SPring-8 X-Ray Free Electron Laser in which a high-intensity short pulse of X-rays interacts with an iron target [11]. The remarkable parameters of the experiment are X-ray energy $h\nu \sim 7.1$ keV, pulse-length 10 fs pulse, focused intensity $I \sim 2 \times 10^{19}$ Watts/cm$^2$. Simple estimates show that during the pulse there are several hundred x-rays per atom in the target.

The handbook cross-section for K-shell absorption by neutral Fe is

$$\sigma_K \sim 2.47 \times 10^{-20} \text{ cm}^2$$

and the lifetime of a K-shell hole is $\tau \sim 1$ fsec. This short lifetime is limited by both X-ray (K $\rightarrow$ L) emission and the Auger effect.

During the pulse, approximately $40\%$ of the Fe atoms should have K-shell holes:

$$\frac{I_{\nu}}{h\nu} \cdot \sigma_K \cdot \tau = 0.39$$

The K-shell absorption edge rises by 600 eV when the same atom has a K-shell hole, so it cannot easily absorb a second photon from the laser source. The absorption edge broadens and shifts when one or more near neighbor atom has a K-shell hole, and such a broadening is visible in the preliminary experimental absorption spectra. Free electrons are also rapidly heated (maybe too rapidly to remain in thermal equilibrium) and this leads to a blurring of the Fermi surface which also permits absorption at lower photon energies.

A many-atom atomic model is the best way to describe this type of experiment because different target atoms are in different ionization/excitation states and different environments during the short pulse of X-rays. A calculation treating the target as having some "temperature" would not capture the inhomogeneity and transient dynamics.

6. Summary and Conclusions

This paper has described a type of atomic-scale simulation which can be called Multi-physics Molecular Dynamics. The method can give atomic-scale models for fusion and other atomic processes.

A key challenge for describing realistic atomic-scale dynamics is to incorporate quantum phenomena (especially abrupt transitions) within a largely classical simulation framework. An interesting method to describe these transitions in treatments of chemical reactions during atomic and/or molecular collisions was proposed by Tully and Preston, [22]. In this work the Landau-Zener approximation was used to justify treating the transition as happening at a specific curve-crossing point in the classical motion.

We would like to find a more general method, which can decide how to change the description from classical (to or from a quantum description) at a predetermined position or time without assuming there is a curve-crossing or other physical transition simultaneous with the change of description. The "small-ball" method summarized in Appendix B gives only a hint of the desired general method.

This method of multiphysics molecular dynamics could be especially useful for preliminary evaluation of new ideas for exotic phenomena. For example, heavy-ion fusion, fusion with spin-polarized fuel, or "ion-quiver" fusion have been proposed but are not being studied experimentally for economic reasons. Particle simulation can offer a first test of these ideas which would help identify candidates for further exploration.

The molecular dynamics simulations can also test the theory of plasma processes by examining situations in which many processes occur simultaneously. The physical models assumed in hydrodynamic simulation codes can only benefit as a result of this type of comparison and testing.
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Appendix A. "Guiding" the Monte Carlo

In the fusion calculations, the fusion probability per DT collision is very small, ~ 10^-6. When the Monte Carlo test is passed, the resulting real fusions are implemented but are at most a few hundred events in 10-20 psec. The fluctuations in this number are about ± 10%.

We also calculate virtual fusions using an enhanced cross-section \( F_{\text{enh}} \times \sigma_{\text{DT}} \). The enhancement factor \( F_{\text{enh}} \) is typically 100. These virtual fusions have the same statistical distribution as a function of particle energy because the enhanced cross-section has the same energy-dependence as the real cross-section. If we implemented these fusions and allowed them to produce energy or deplete the fuel, it would dramatically alter the ignition dynamics. However we can use them to improve the statistics of the calculation because they have smaller fluctuations (~ 1%).

We use the virtual fusions to predict the output neutron time-of-flight spectrum and obtain a smooth spectrum consistent with the spectrum from the real fusions. However, the ratio

\[
\frac{\text{Number of virtual fusions}}{F_{\text{enh}}}
\]

also predicts the number of real fusions with less fluctuation.

It is possible to dynamically adjust the fusion conditional probability used in the MC test to guide the number of real fusions toward the expectation based on the virtual fusions. When we do this we can obtain million-particle accuracy from a 10,000-particle calculation. We used this method in re-runs of some of our reference cases and find the algorithm has entirely satisfactory behavior.

Appendix B. "Small-ball" method

In the OK code, we treat ion-ion Coulomb collisions by analytic solution rather than by numerical integration. This is combined with a MC test for QM processes such as fusion (or radiation emission in previous versions of the code).

Some advantages of this small-ball method [15, 23, 24] are: The method makes it easy to calculate radiation emission and absorption consistent with detailed balance. The method gives a way to calculate fusion reactions and quantum transitions in atomic kinetics. The calculation assumes a pure Coulomb potential inside the small ball, with no need for a pseudopotential. The small-ball method allows use of large time-steps for the MD calculation.

Some difficulties of the method are, in brief: it must be integrated into MD code and this raises issues about the time-steps, synchronization, labeling and storage issues. The small-ball method neglects some forces between ions for short times (while they're small), and there is a rare possibility of multiple ions being inside the small ball at the same time.

For fusion reactions, the small-ball approximation assigns a conditional probability of reaction

\[
P_{\text{Cond}} = \frac{\sigma_{\text{reaction}}}{\pi R_{\text{sb}}^2}
\]

(B-1)

to be tested each time that a D and T ion collide. Since the arriving flux is proportional to \( \pi R_{\text{sb}}^2 \), the reaction rate is approximately independent of \( R_{\text{sb}} \).

As a more general question, we can argue that when a particle crosses a predefined surface (such as the surface of a small sphere), we want to change description from a classical
description with position-momentum coordinates \( r, p \) to a quantum description with a wave-function \( \psi_j \). This change should be governed by a conditional probability \( P_{\psi_j}(\psi_j) \) to decide which wave function is chosen in each individual case.

How should this conditional probability be chosen to obtain the most realistic description of the particle dynamics? Conversely, when the particle leaves the sphere, it should again select the outgoing coordinates \( r', p' \) using another conditional probability \( P_{\psi_j}(r', p') \). What is the best way to select these conditional probabilities?

We can propose use of Bayes theorem and the Wigner distribution to guide these choices. At least in a near-equilibrium system, we have \textit{a priori} probabilities \( P(r, p) \) and \( P(\psi_j) \) for the classical and quantum probabilities; these can be obtained from Maxwell or Fermi-Dirac statistics, depending on the system. The conditional probabilities should then obey Bayes' theorem,

\[
P(r, p) P_{\psi_j}(\psi_j) = P(\psi_j) P_{\psi_j}(r, p)
\]  

(B-2)

In this equation we can use the Wigner distribution,

\[
P_{\psi_j}(r_o, p_o) = \left\langle \psi_j \left| \delta(r - r_o) \delta(p - p_o) \right| \psi_j \right\rangle
\]  

(B-3)

to determine the conditional probability for \( r_o, p_o \) given the wave-function \( \psi_j \). (This expression should be symmetrized by taking the real part.) If we know the wave function in both position-space \( \psi_j(r_o) \) and momentum-space \( \phi_j(p_o) \), then the Wigner distribution can be explicitly evaluated as

\[
\left\langle \psi_j \left| \delta(r - r_o) \delta(p - p_o) \right| \psi_j \right\rangle = \psi_j^*(r_o) \frac{e^{ip_o \cdot p}}{(2\pi)^{3/2}} \psi_j(p_o)
\]  

(B-4)

This method has the well-known imperfection that the Wigner distribution assigns unphysical negative probabilities over a small part of the \( r, p \) phase-space.

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