Molecular dynamics simulations of strongly coupled plasmas

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Molecular dynamics simulations of strongly coupled plasmas

Results obtained in collaboration with:

- G. J. Kalman - Boston College, USA
- P. Hartmann - RISSP Budapest, Hungary
- K. I. Golden - University of Vermont, USA
- J. Goree - University of Iowa, USA

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Outline

- Why do we need simulations?
- Systems of interest
- Basics of Molecular Dynamics (MD) simulations
- What do we learn from MD?
- Structural & thermodynamic properties
- Localization and transport
- Collective excitations
Why do we need simulations?

- **Simulations are useful**
  - for checking theoretical results
  - for cases where no theoretical results are available
  - for understanding experimental observations

- **Simulations allow**:
  - identification of important processes
  - visualization of the system

- *Most dramatic advance of resources is experienced in the field of simulations*
Dramatic advance of resources

Monte Carlo Study of a One-Component Plasma. I*

S. G. BRUSH†
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AND

H. L. SAHLIN AND E. TELLER
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(Received 28 March 1966)

been made of a plasma of heavy ions immersed in a uniform neutralizing back-ground from 32 to 500 particles, with periodic boundary conditions, were used. The results in terms of a dimensionless parameter $\Gamma = (4\pi n/3)^{1/3} (Ze)^{1/3} T^{1/3}$, where $n$ is the number density per cubic centimeter, $T$ is the temperature (degrees Kelvin), $Z$ is the ionic charge, and $E$ is the atomic number. Thermodynamic properties were obtained for values of $\Gamma$ ranging from 0.0 to 100. (MC) method.

Pioneering MD simulations in the 1970s-80s (OCP, BIM, statics, dynamics, transport, etc.)
Where a calculator on the ENIAC is equipped with 18,000 vacuum tubes and weighs 30 tons, computers in the future may have only 1,000 vacuum tubes and weigh only 1.5 tons.

[Popular Mechanics, 1949]
Systems of interest

Classification of plasmas from the American Physical Society “fusion chart”

Is there really not much here to look for ???
Plasmas…. a better phase diagram

Consider the interaction between a single type of particles (ion-ion)

\[ \Gamma = \frac{V_{\text{POT}}}{V_{\text{KIN}}} \]

\( \Gamma = 1 \)

STRONGLY COUPLED PLASMAS

The one-component plasma (OCP) model: only one type of species is considered explicitly, the presence and effects of other species are accounted for by the potential.

**Coulomb**

Coulomb potential:
\[
\Phi(r) = \frac{1}{4\pi\varepsilon_0} \frac{Q}{r}
\]
non-polarizable background

**Debye-Hückel / Yukawa**

D-H / Yukawa potential & screening parameter:
\[
\Phi(r) = \frac{1}{4\pi\varepsilon_0} \frac{Q \exp(-r/\lambda_D)}{r}
\]
polarizable background

**Characteristic energies** (Coulomb):
\[
V_{\text{POT}} = \frac{Q^2}{4\pi\varepsilon_0 a} \quad V_{\text{KIN}} = kT
\]
a : Wigner-Seitz radius

**Coupling parameter**:
\[
\Gamma = \frac{V_{\text{POT}}}{V_{\text{KIN}}} = \frac{1}{4\pi\varepsilon_0} \frac{Q^2}{akT}
\]
Molecular Dynamics (MD) basics
(one-component plasma ⇒ strongly interacting classical many-body system)

Equilibrium & non-equilibrium MD

- We let the system evolve according to interactions
- Perturb the system and measure response
Molecular Dynamics (MD) simulation basics

**Equilibrium MD**

**SIMULATION CORE + MEASUREMENTS**

Time evolution of phase space trajectories of an ensemble of $N$ particles

Calculate quantities of interest from phase space coordinates

Example: finite system with external confinement:

$$m\ddot{r}_i = \sum_{i\neq j} F_{i,j}(t) + F_{\text{ext}}(t) - m\eta v_i(t) + R$$

- Friction
- Brownian randomly fluctuating force (Langevin force)

$$F_{i,j} = -\frac{\partial \phi(r_{ij})}{\partial r}$$

$$F_{\text{ext}} = -fr^2 \text{ (e.g.)}$$
Molecular Dynamics (MD) simulation basics

Integration of the equation of motion ("leapfrog scheme")

\[
\begin{align*}
\mathbf{v}_i\left(t - \frac{\Delta t}{2}\right) & \quad \mathbf{r}_i(t) \quad \mathbf{v}_i\left(t + \frac{\Delta t}{2}\right) \quad \mathbf{r}_i(t + \Delta t) \\
\mathbf{v}_i\left(t + \frac{\Delta t}{2}\right) & = \mathbf{v}_i\left(t - \frac{\Delta t}{2}\right) + \frac{\mathbf{F}_i(t)}{m} \Delta t \\
\mathbf{r}_i(t + \Delta t) & = \mathbf{r}_i(t) + \mathbf{v}_i\left(t + \frac{\Delta t}{2}\right)
\end{align*}
\]

How to calculate \( \sum F_{i,j}(t) \) ?
Molecular Dynamics (MD) simulation basics

Short – range interaction potentials
Interaction is considered only between “closely-separated” pairs of particles (cutoff radius)

\[ F_i(t) = \sum_{r_{ij} < r_C} F_{i,j}(t) \]
Molecular Dynamics (MD) simulation basics

Long – range interaction potentials (e.g. Coulomb):

Not possible to find cutoff radius, “tricks” are needed

\[ F_i(t) = \sum_{\text{cell+images}} F_{i,j}(t) \]

Possible solutions:
- Ewald summation
- Particle-Particle, Particle-Mesh (PPPM, P3M) method (Hockney & Eastwood)
Molecular Dynamics (MD) simulation basics

The PPPM method uses finite size charge clouds

$$\rho(r) = \rho_0 \left(1 - \frac{r}{R}\right)$$

$$\int_0^R \rho(r) dV = Q$$

Fourier transform is band-limited, the interaction between clouds can be represented on a mesh in \(k\)-space, images are included (PM)

if \(r \geq R\):

$$F(\cdot \cdot \cdot) = F(\cdot \cdot \cdot)$$

if \(r < R\):

$$F(\cdot \cdot \cdot) = F(\cdot \cdot \cdot) + F_{\text{corr}}(r)$$


Correction force, to be applied for closely separated neighbors only (PP, chaining mesh)
Molecular Dynamics: What do we see?

$\Gamma = 120, \kappa = 1$

$\Gamma = 5, \kappa = 1$

2D frictionless Yukawa liquids
Molecular Dynamics: What do we learn?

- Phase space coordinates \((r_i, v_i) \; i = 1 \ldots N\)
- Correlation functions
- Identification of collective modes
- Calculation of transport parameters
- Structure
- Thermodynamic quantities

\[ (r_i, v_i) \; i = 1 \ldots N \]
It’s real: experimental realization of 2D dusty plasma

- Dust particles dispersed in a glow discharge plasma acquire a charge of $\sim 10^4 q_e$
- Dust layer is levitated due to the balance between electrostatic force and gravity
- Interaction: screened Coulomb (Yukawa) potential
- Crystallization at high $\Gamma$
- Quasi-2D confinement
- Extensive experimental work from early 1990s (Morfill, Thomas, Goree, Fortov, Piel, et al., ......) in the crystal and liquid phases

$\Phi(r) = \frac{1}{4\pi\varepsilon_0} \frac{Q \exp(-r/\lambda_D)}{r}, \quad \kappa = \frac{a}{\lambda_D}$
Experimental realization of 2D dusty plasma

Dusty plasma experiment in RISSP, Budapest (P. Hartmann)

melamine-formaldehyde microspheres
Experimental realization of 2D dusty plasma

Laser manipulation to measure complex viscosity

Structural and thermodynamic properties
Pair correlation & thermodynamic properties

Pair correlation function

- Energy:
  \[ \frac{E}{N} = \frac{3}{2} k_B T + \frac{n}{2} \int_0^\infty \varphi(r)g(r) \, 4\pi r^2 \, dr \]

- Pressure:
  \[ p = nk_B T - \frac{n^2}{6} \int_0^\infty \frac{\partial \varphi(r)}{\partial r} g(r) \, 4\pi r^3 \, dr \]

- Isothermal compressibility:
  \[ k_B T \left( \frac{\partial n}{\partial p} \right)_T = 1 + n \int_0^\infty [g(r) - 1] \, 4\pi r^2 \, dr \]

E.g. 3D Coulomb OCP (one-component plasma)

Strong correlation, liquid-like structure at high coupling
Phase transitions:
3D Coulomb / Yukawa systems

**Coulomb**
(Monte Carlo)

S. G. Brush, H. L. Sahlin and E. Teller,

$\Gamma \approx 125$

E. L. Pollock and J. P. Hansen

G. S. Stringfellow, H. E. DeWitt and W. L. Slattery,

$\Gamma \approx 175$

**Yukawa**

S. Hamaguchi, R.T. Farouki and D.H.E. Dubin,

$\Gamma$
Transport phenomena

Diffusion, shear viscosity and thermal conductivity in 3D systems
(although 2D is more interesting 😊)
Measurements of transport coefficients

Equilibrium Molecular Dynamics:

Measure correlation functions

\[
D = \frac{1}{2} \int_0^\infty C_v \, dt \\
\eta = \frac{1}{V kT} \int_0^\infty C_\eta \, dt \\
\lambda = \frac{1}{V kT^2} \int_0^\infty C_\lambda \, dt
\]

\[C_v \equiv \langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle \quad \text{VACF}\]

\[C_\eta \equiv \langle P_{xy}(t) P_{xy}(0) \rangle \quad \text{SACF}\]

\[C_\lambda \equiv \langle J_{Qx}(t) J_{Qx}(0) \rangle \quad \text{EACF}\]

Non-Equilibrium Molecular Dynamics:

Perturb the system and measure the response
Diffusion coefficient

\[ D^* = \frac{D}{a^2 \omega_p} \]


\[ D^* = \alpha(T^* - 1)\beta + \gamma \]
\[ T^* = \frac{T}{T_M} \]

Universal scaling:

Shear viscosity: methods (1)

\[ P_{xy} = \sum_{i=1}^{N} \left[ m v_{ix} v_{iy} - \frac{1}{2} \sum_{j \neq i}^{N} \frac{x_{ij} y_{ij}}{r_{ij}} \frac{\partial \phi(r_{ij})}{\partial r_{ij}} \right] \]

\[ \eta = \frac{1}{V kT} \int_{0}^{\infty} \langle P_{xy}(t) P_{xy}(0) \rangle dt \]

\[ W(y_k) = W_{M0} \sin \left( \frac{2\pi y_k}{L} \right) \]

\[ \frac{\partial v_x}{\partial t} = \frac{\eta}{\rho} \frac{\partial^2 v_x}{\partial y^2} \]

\[ W(y, t) = W_{M0} \sin \left( \frac{2\pi y}{L} \right) \exp \left( -\frac{t - t_0}{\tau} \right) \]

\[ \eta = \frac{\rho}{\tau} \left( \frac{L}{2\pi} \right)^2 \]

Z. Donkó and B. Nyíri, Phys. Plasmas 7, 45 2000

Shear viscosity: methods (2)

Reverse Molecular Dynamics

**External momentum transfer**

\[
\eta \frac{dv_x(y)}{dy} = \frac{\Delta p}{2t_{\text{sim}} S}
\]


Homogeneous Shear Algorithm

\[
\frac{dx_i}{dt} = \tilde{p}_i + \gamma y_i \hat{x} \\
\frac{d\tilde{p}_i}{dt} = F_i - \gamma \tilde{p}_{yi} \hat{x} - \alpha \tilde{p}_i
\]

\[
\eta = - \lim_{t \to \infty} \frac{\langle P_{xy}(t) \rangle}{\gamma}
\]

Shear viscosity of 3D Coulomb liquids

\[ \eta' = \eta \frac{m n \omega_p a^2}{\Gamma} \]

Equilibrium MD:
- B. Bernu, P. Vieillefosse, and J. P. Hansen, Phys. Lett. A 63, 301 (1977);
- J. Daligault, Phys. Rev. Lett. 96, 065003 (2006) (Scaled) high \( \Gamma \) Arrhenius fit
- G. Salin and J.-M. Caillol, Phys. Rev. Lett. 88, 065002 (2002);
- G. Salin and J.-M. Caillol, Phys. Plasmas 10, 1220 (2003) (\( \kappa = 0.01 \))
- T. Saigo and S. Hamaguchi, Phys. Plasmas 9, 1210 (2002) (\( \kappa = 0.1 \))

Transient perturbation MD:
Thermal conductivity: MD methods

Reverse molecular dynamics

Spatial temperature modulation

\[
\frac{\partial T}{\partial t} = \frac{\lambda}{c \rho} \frac{\partial^2 T}{\partial x^2}
\]

\[
\lambda = \frac{c \rho}{\tau_H} \left( \frac{L}{2 \pi} \right)^2
\]

\[
\lambda = \frac{\Delta E}{2 S t_{\text{sim}} \langle \frac{\Delta T}{\Delta x} \rangle}
\]

F. Müller-Plathe,

Z. Donkó, B. Nyíri, L. Szalai, and S. Holló,
Thermal conductivity of 3D Coulomb liquids

\[ \lambda' = \frac{\lambda}{nk\omega_p a^2} \]

\[ \text{Slope} = \frac{1}{\tau_H} \]

\[ x/L \]

\[ t [\text{ps}] \]

\[ t [\text{ps}] \]

\[ \ln(T_m) \]


N=8192

N=1024


N=128 ... 864, \( \kappa = 0.01 \)


N=6400, \( \kappa = 0.1 \)

N=1600, \( \kappa = 0.1 \)
Collective excitations
Collective excitations in 3D plasma liquids

Microscopic density fluctuations:
\[ \rho(k, t) = \sum_{j=1}^{N} \exp[i k x_j(t)] \]

Dynamical structure function:
\[ S(k, \omega) = \frac{1}{2\pi N} \lim_{\Delta T \to \infty} \frac{1}{\Delta T} |\rho(k, \omega)|^2 \]
\[ \rho(k, \omega) = \mathcal{F}[\rho(k, t)] \]

Microscopic current fluctuations:
\[ \lambda(k, t) = \sum_{j=1}^{N} v_{jx}(t) \exp[i k x_j(t)] \]

Longitudinal and transverse current-current fluctuation spectra
\[ L(k, \omega) \quad T(k, \omega) \]

\[ \tau(k, t) = \sum_{j=1}^{N} v_{jy}(t) \exp[i k x_j(t)] \]
Collective excitations in 3D plasma liquids

Coulomb:
- L : const. freq.
- T : acoustic

Yukawa:
- L : quasi-acoustic
- T : acoustic
Collective excitations in 3D liquids: MD vs. theory

\[ \Lambda_{3D}(x, y) = -2 \frac{e^{-y}}{x} \left[ (1 + y + y^2) \left( \frac{\sin(x)}{x} + 3 \frac{\cos(x)}{x^2} - 3 \frac{\sin(x)}{x^3} \right) - \frac{y^2}{6} \left( 1 + 3 \frac{\sin(x)}{x} + 12 \frac{\cos(x)}{x^2} - 12 \frac{\sin(x)}{x^3} \right) \right] \]


QLCA theory:

\[ \Omega_L^2(k) = \Omega_0^2(k) + \omega_{0,3D}^2 \frac{\bar{k}^2}{2} \int_0^\infty \Lambda_{3D}(\bar{k}\bar{r}, \kappa\bar{r}) h(\bar{r}) d\bar{r} \]

\[ \Omega_T^2(k) = \omega_{0,3D}^2 \frac{\bar{k}^2}{2} \int_0^\infty \Theta_{3D}(\bar{k}\bar{r}, \kappa\bar{r}) h(\bar{r}) d\bar{r} \]

Cutoff of T-mode at finite \( k \): liquid

M.S. Murillo

EXP: J. Goree

\[ \Omega_0^2(k) = \omega_{0,3D}^2 \frac{\bar{k}^2}{\bar{k}^2 + \kappa^2} \]
Simulation studies aid the understanding of theoretical and experimental results.

Simulations are suitable for a wide variety of strongly coupled many-particle systems.

Equilibrium / non-equilibrium Molecular Dynamics simulations can be used to study:

- Structural & thermodynamical properties
- Localization and transport properties
- Collective excitations
- ......

Thank you for your attention.