

Molecular dynamics simulations of strongly coupled plasmas



Workshop on Dynamics and Control of Atomic and
Molecular Processes Induced by Intense Ultrashort Pulses
- CM0702 WG2, WG3 meeting -

27-30 September 2011, Debrecen, Hungary

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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

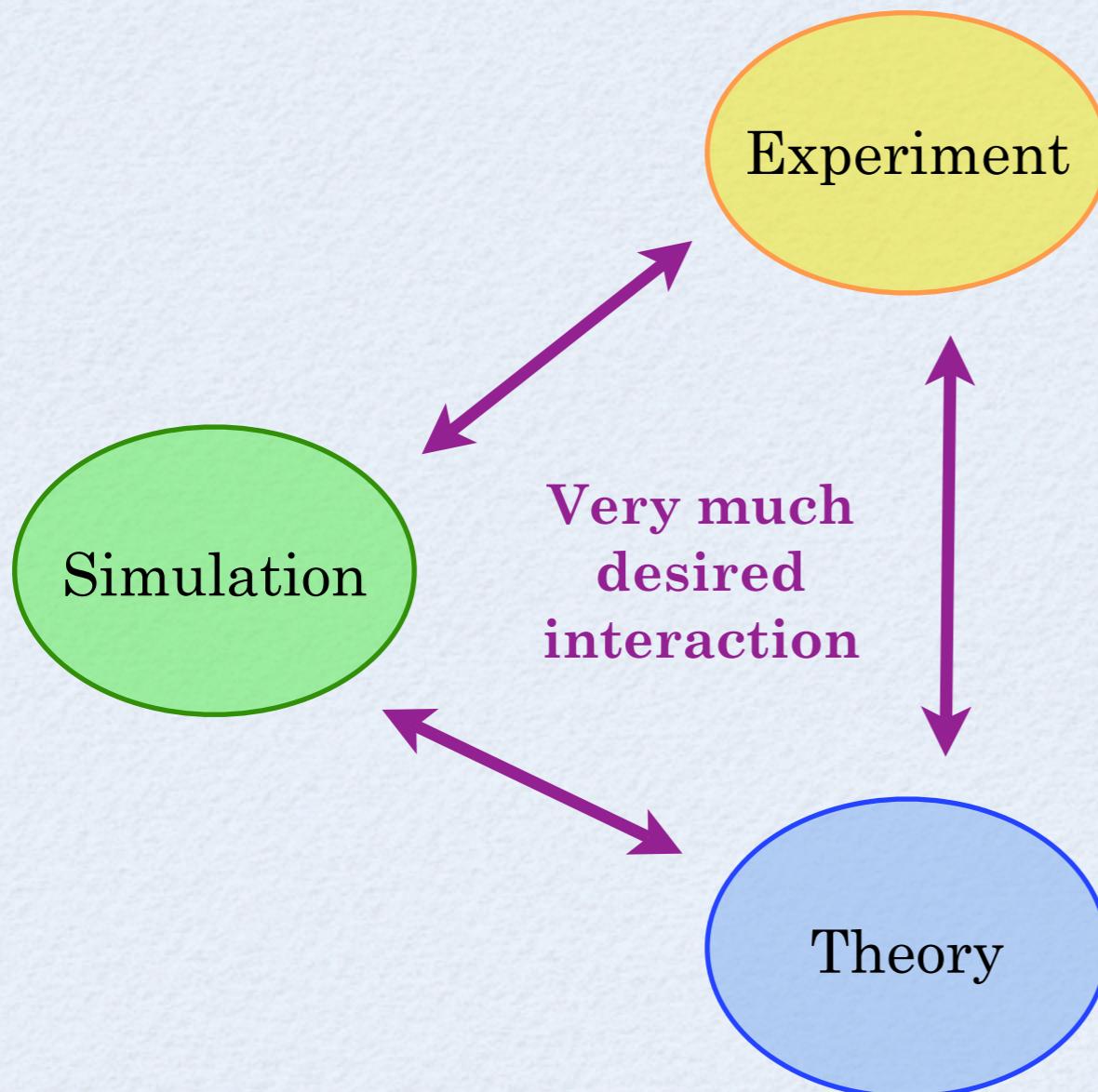
Supported by:

- NSF / MTA (HU Academy of Sciences) / OTKA (HU Sci. Research Fund)

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

THE JOURNAL OF CHEMICAL PHYSICS VOLUME 45, NUMBER 6 15 SEPTEMBER 1966

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

S. G. BRUSH†

Lawrence Radiation Laboratory, University of California, Livermore, California

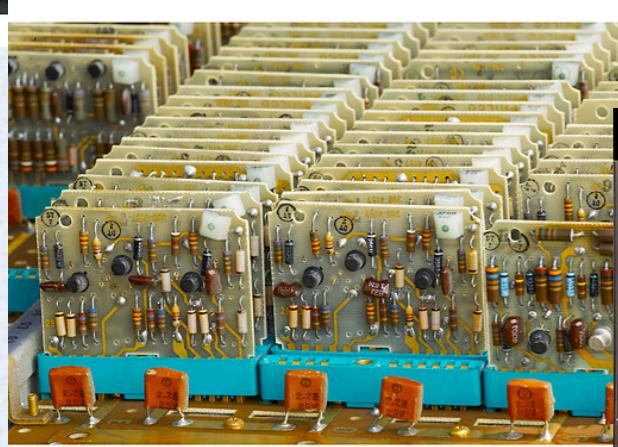
AND

H. L. SAHLIN AND E. TELLER

*Lawrence Radiation Laboratory, University of California, Livermore, California, and
Department of Applied Science, University of California, Davis/Livermore, California*

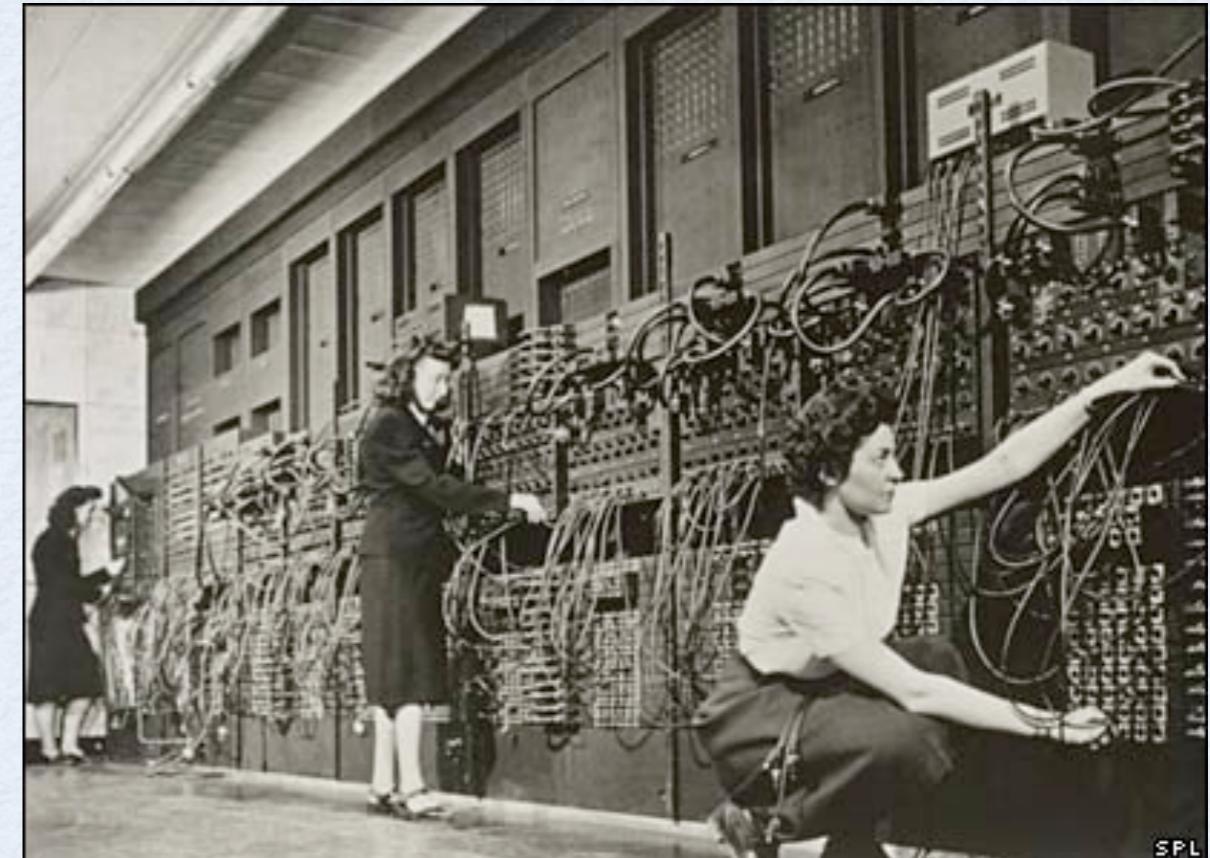
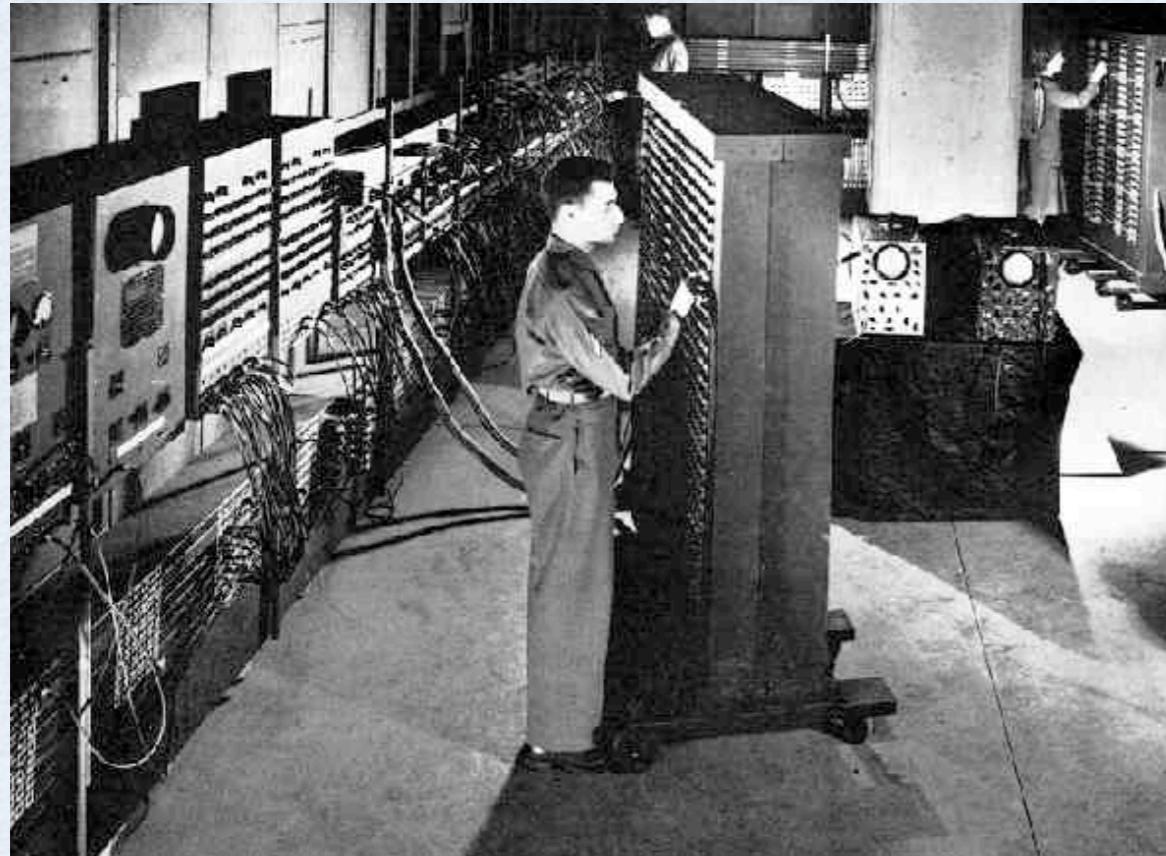
(Received 28 March 1966)

been made of a plasma of heavy ions immersed in a uniform neutralizing background of electrons. Systems containing from 32 to 500 particles, with periodic boundary conditions, were used. The properties of the plasma were determined in terms of a dimensionless parameter $\Gamma = (4\pi n/3)^{1/3} [(Ze)^2/kT]$, where n is the density (particles per cubic centimeter), T is the temperature (degrees Kelvin), e is the electronic charge, and Z is the atomic number. Thermodynamic properties of the plasma were obtained for values of Γ ranging from 0.05 to 100 by the Monte Carlo (MC) method.



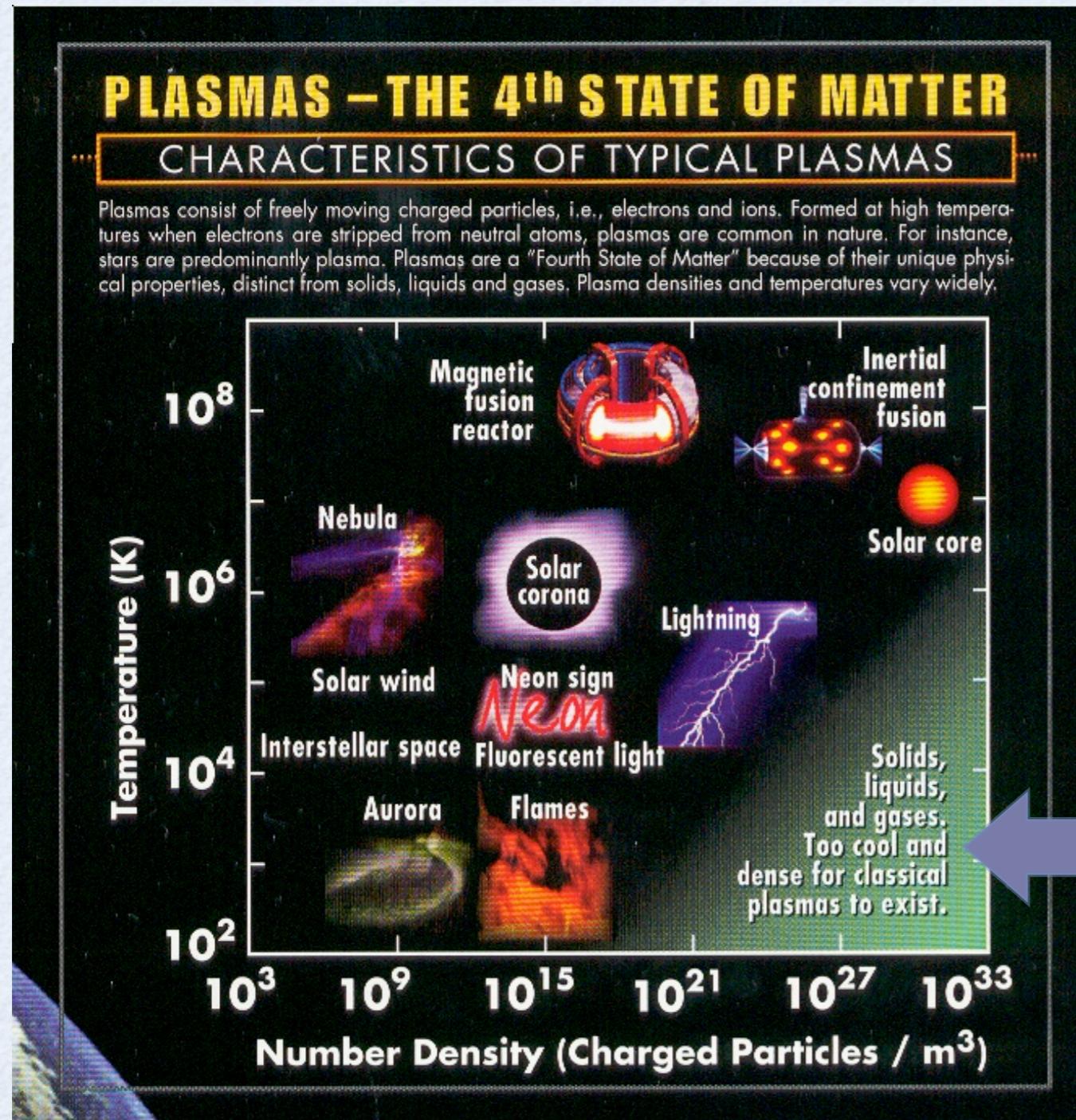
Pioneering MD simulations in the 1970s-80s
(OCP, BIM, statics, dynamics, transport, etc.)

Dramatic advance of resources ... where does this go?



"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]

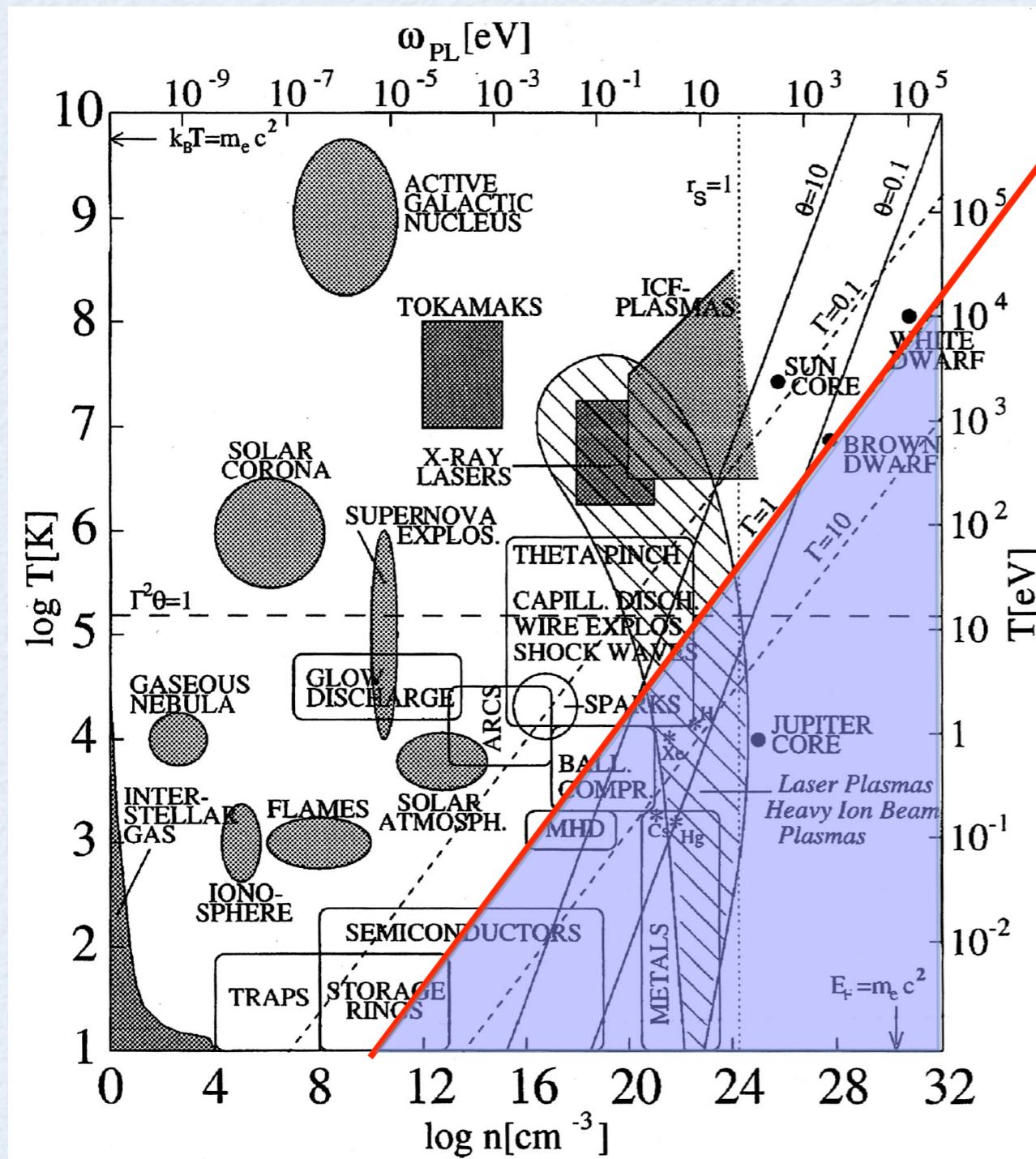
Systems of interest



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

Is there really
not much here
to look for ???

Plasmas.... a better phase diagram



$$\Gamma = 1$$

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

$$\Gamma = \frac{V_{\text{POT}}}{V_{\text{KIN}}}$$

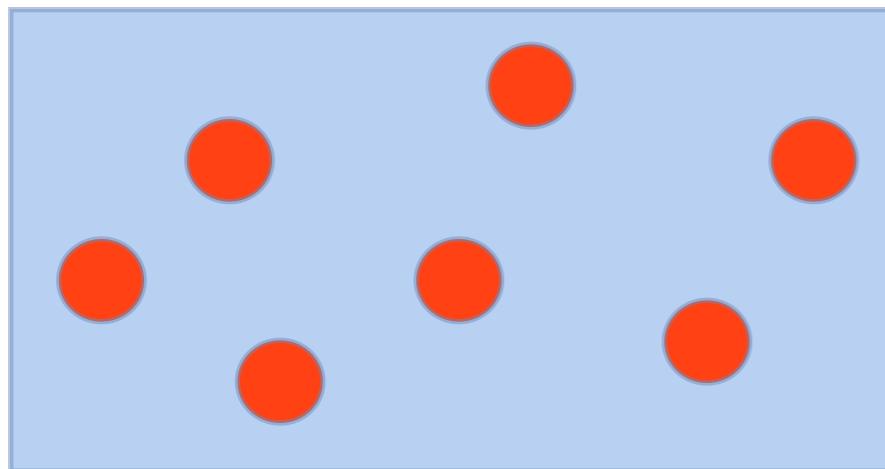
STRONGLY COUPLED PLASMAS

R. Redmer, Phys. Reports 282, 35 (1997)

The one-component plasma (OCP) model

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:

$$\Phi(r) = \frac{1}{4\pi\epsilon_0} \frac{Q}{r}$$

non-polarizable
background

Characteristic energies
(Coulomb):

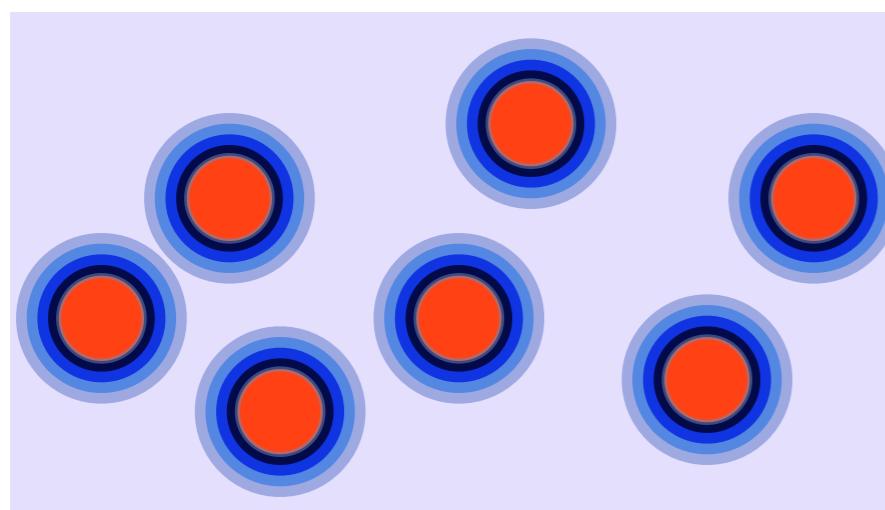
$$V_{\text{POT}} = \frac{Q^2}{4\pi\epsilon_0 a} \quad V_{\text{KIN}} = kT$$

a : Wigner-Seitz radius

Coupling parameter:

$$\Gamma = \frac{V_{\text{POT}}}{V_{\text{KIN}}} = \frac{1}{4\pi\epsilon_0} \frac{Q^2}{akT}$$

Debye-Hückel / Yukawa



D-H / Yukawa potential & screening parameter:

$$\Phi(r) = \frac{1}{4\pi\epsilon_0} \frac{Q \exp(-r/\lambda_D)}{r} ,$$

$$\kappa = \frac{a}{\lambda_D}$$

polarizable background

Molecular Dynamics (MD) basics

(one-component plasma \Rightarrow 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

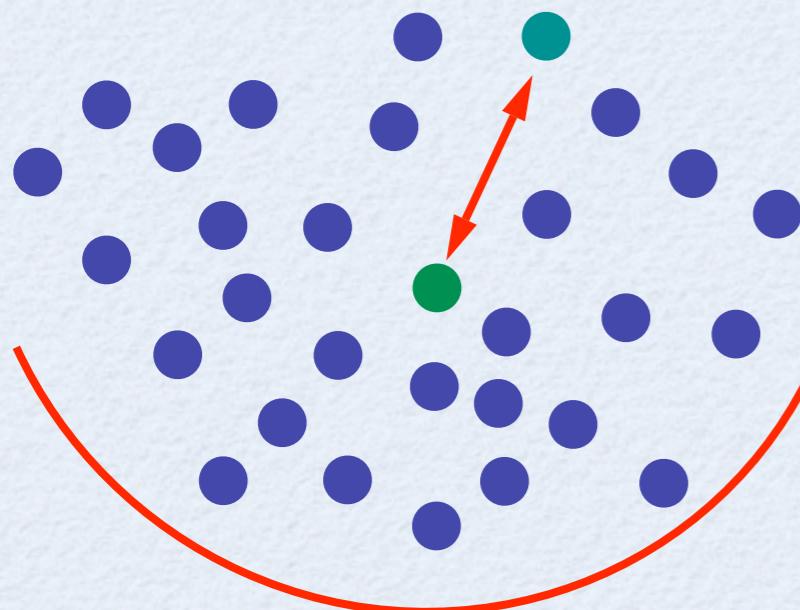
Equilibrium MD

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:



$$m\ddot{\mathbf{r}}_i = \sum_{i \neq j} \mathbf{F}_{i,j}(t) + \mathbf{F}_{\text{ext}}(t) - m\eta\mathbf{v}_i(t) + \mathbf{R}$$

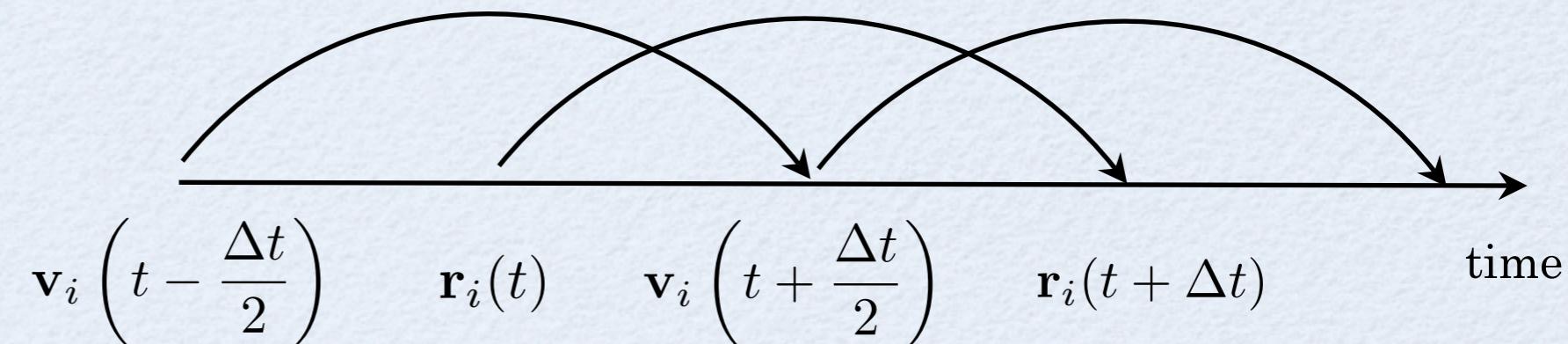
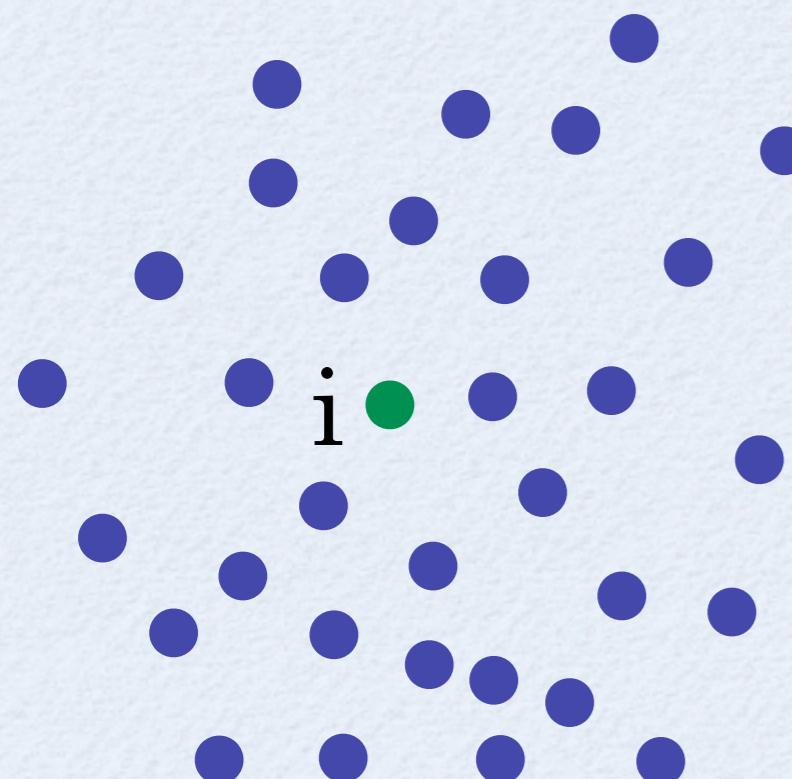
$$\mathbf{F}_{i,j} = -\frac{\partial\phi(r_{ij})}{\partial r}$$

$$\mathbf{F}_{\text{ext}} = -fr^2 \text{ (e.g.)}$$

Friction
Brownian randomly fluctuating force (Langevin force)

Molecular Dynamics (MD) simulation basics

Integration of the equation of motion (“leapfrog scheme”)



$$\mathbf{v}_i \left(t - \frac{\Delta t}{2} \right) \quad \mathbf{r}_i(t) \quad \mathbf{v}_i \left(t + \frac{\Delta t}{2} \right) \quad \mathbf{r}_i(t + \Delta t)$$

$$\mathbf{v}_i \left(t + \frac{\Delta t}{2} \right) = \mathbf{v}_i \left(t - \frac{\Delta t}{2} \right) + \frac{\mathbf{F}_i(t)}{m} \Delta t$$

$$\mathbf{r}_i(t + \Delta t) = \mathbf{r}_i(t) + \mathbf{v}_i \left(t + \frac{\Delta t}{2} \right)$$

$$m\ddot{\mathbf{r}}_i = \sum_{i \neq j} \mathbf{F}_{i,j}(t) + \mathbf{F}_{\text{ext}}(t) - m\eta\mathbf{v}_i(t) + \mathbf{R}$$

How to calculate $\sum \mathbf{F}_{i,j}(t)$?

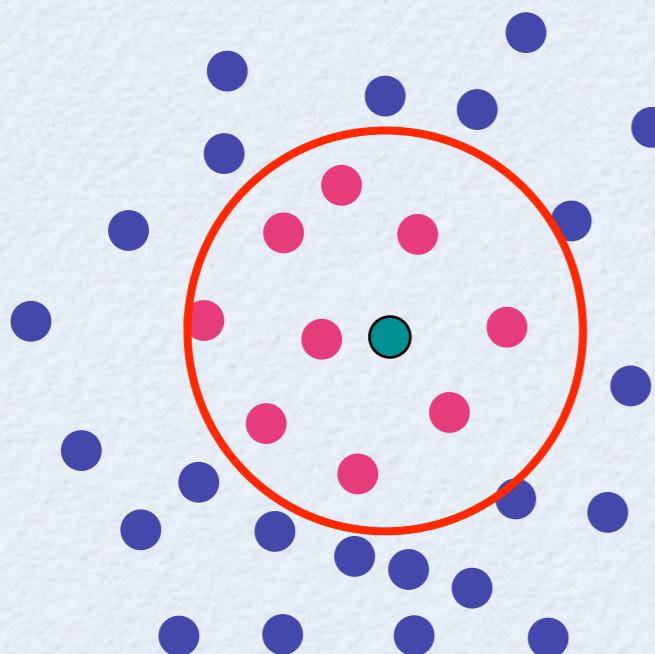
Molecular Dynamics (MD) simulation basics

Short – range interaction potentials

Interaction is considered only between “closely-separated” pairs of particles (cutoff radius)

$$\mathbf{F}_i(t) = \sum_{r_{ij} < r_C} \mathbf{F}_{i,j}(t)$$

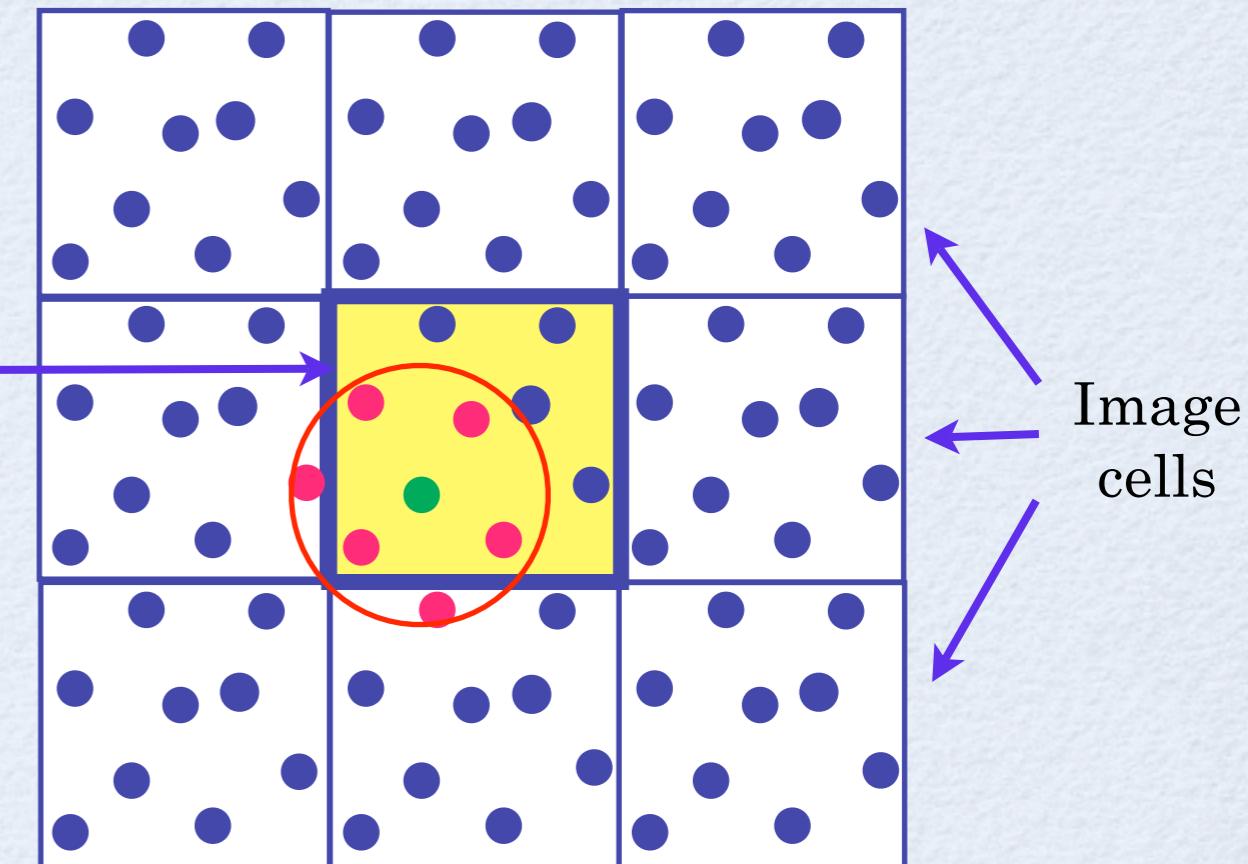
Finite system



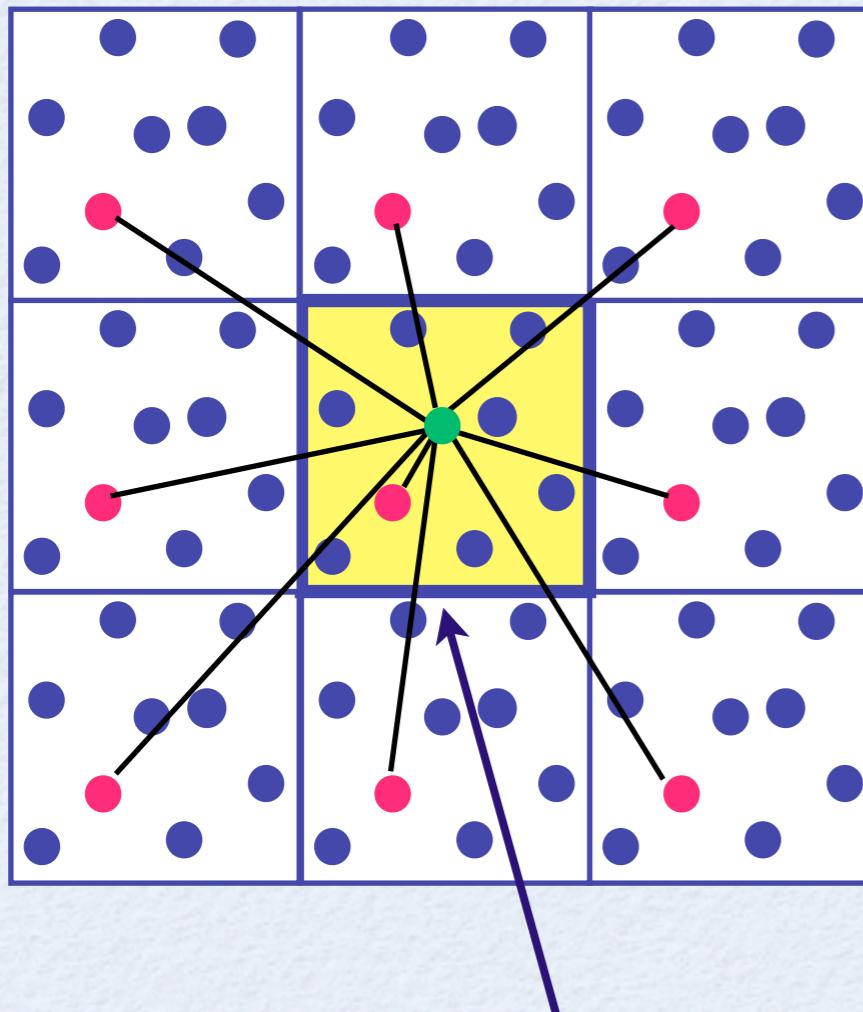
Primary
simulation cell

Infinite system

PERIODIC BOUNDARY CONDITIONS



Molecular Dynamics (MD) simulation basics



Primary simulation cell
(yellow)

Long – range interaction potentials

(e.g. Coulomb):

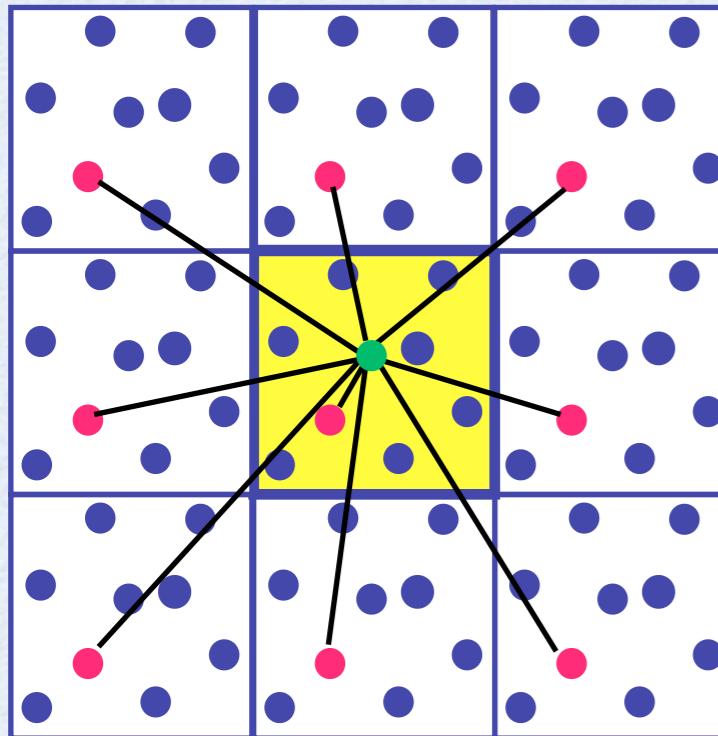
Not possible to find cutoff radius,
“tricks” are needed

$$\mathbf{F}_i(t) = \sum_{\text{cell+images}} \mathbf{F}_{i,j}(t)$$

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 \mathbf{k} -space, images are included (PM)

$$\text{if } r \geq R : F(\bullet \bullet) = F(\bullet \bullet)$$

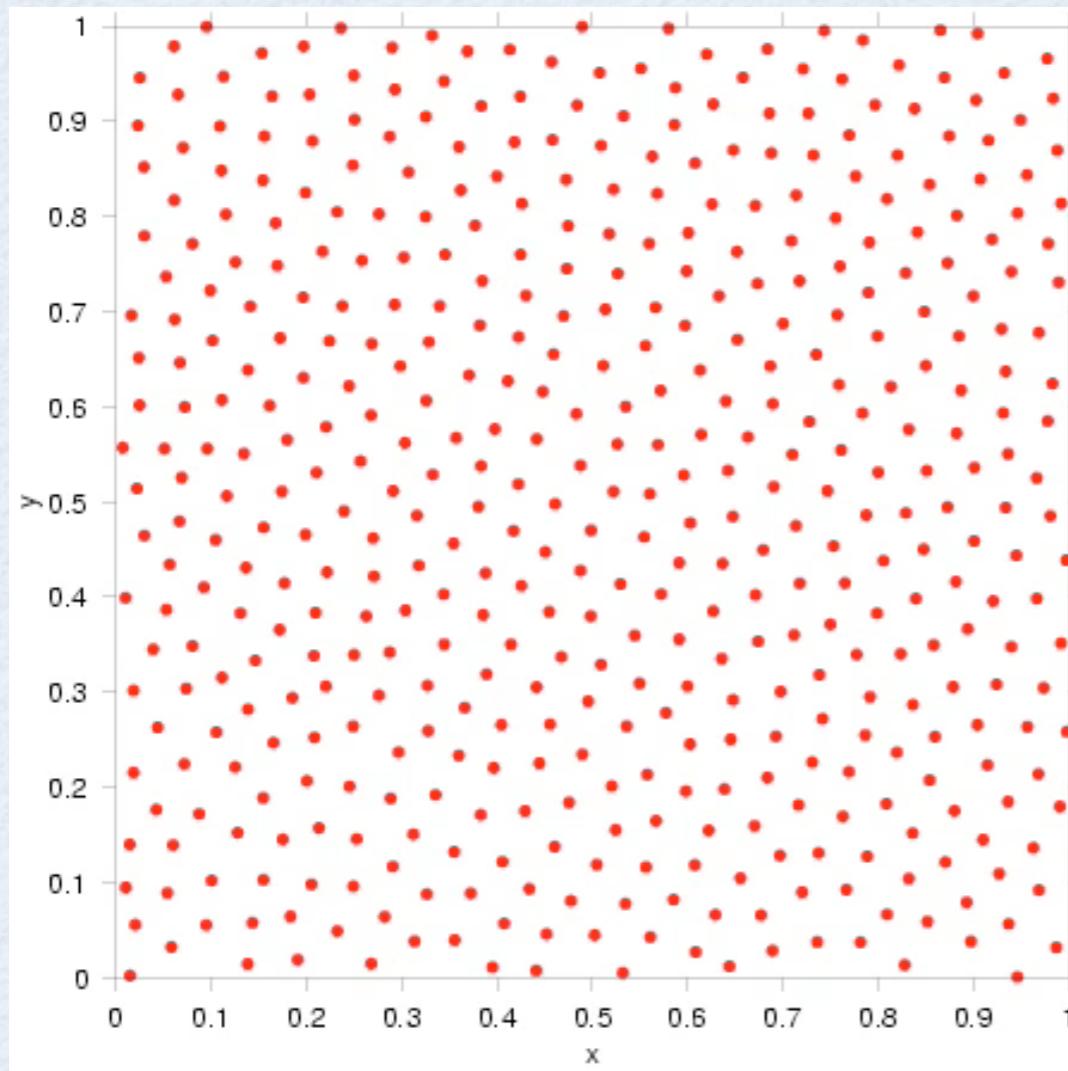
$$\text{if } r < R : F(\bullet \bullet) = F(\bullet \bullet) + F_{\text{corr}}(r)$$

Hockney R W and Eastwood J W 1981
Computer Simulation Using Particles
(New York: McGraw-Hill)

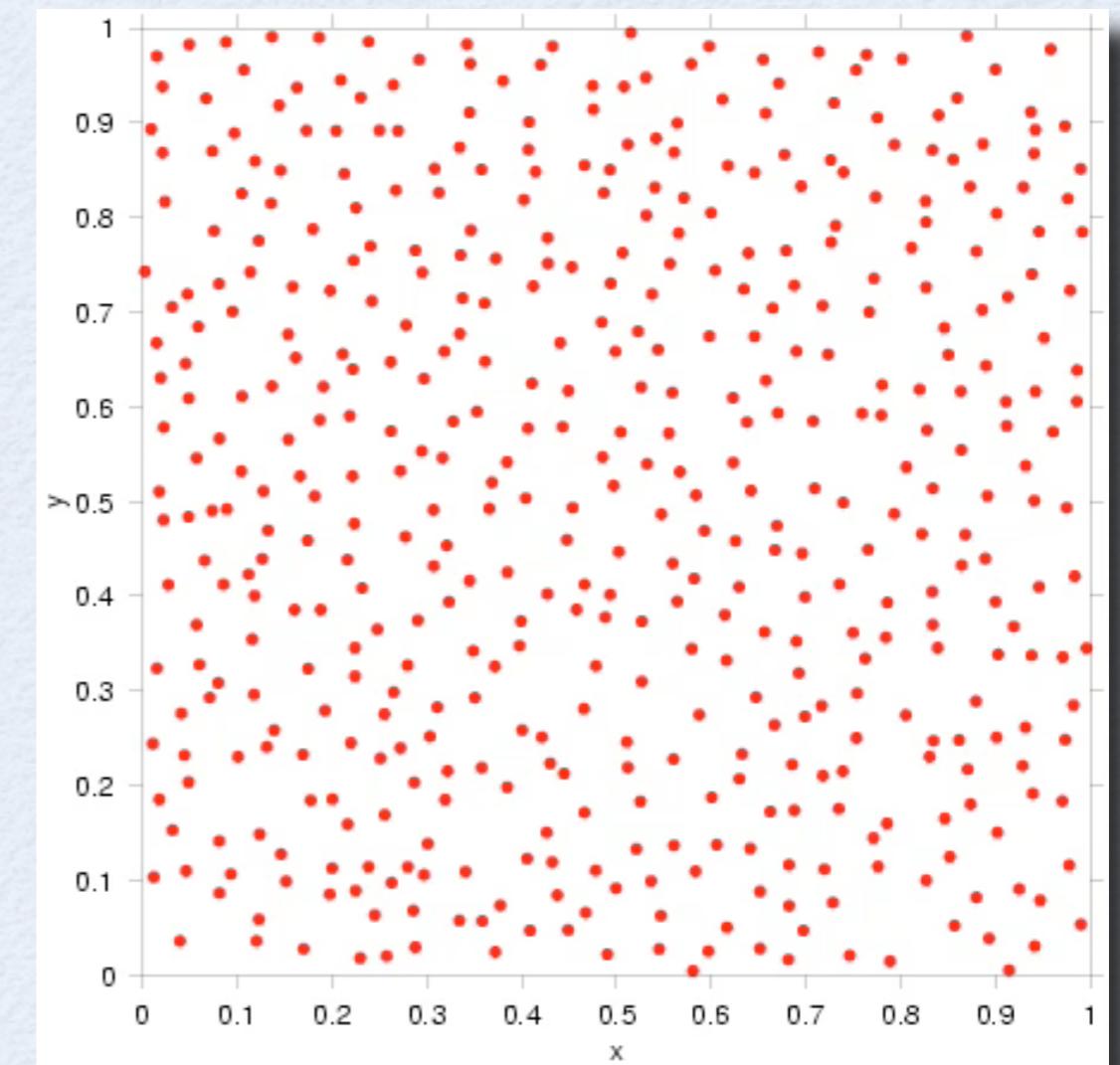
Correction force, to be applied for closely separated neighbors only (PP, chaining mesh)

Molecular Dynamics : What do we see?

$\Gamma=120, \kappa=1$

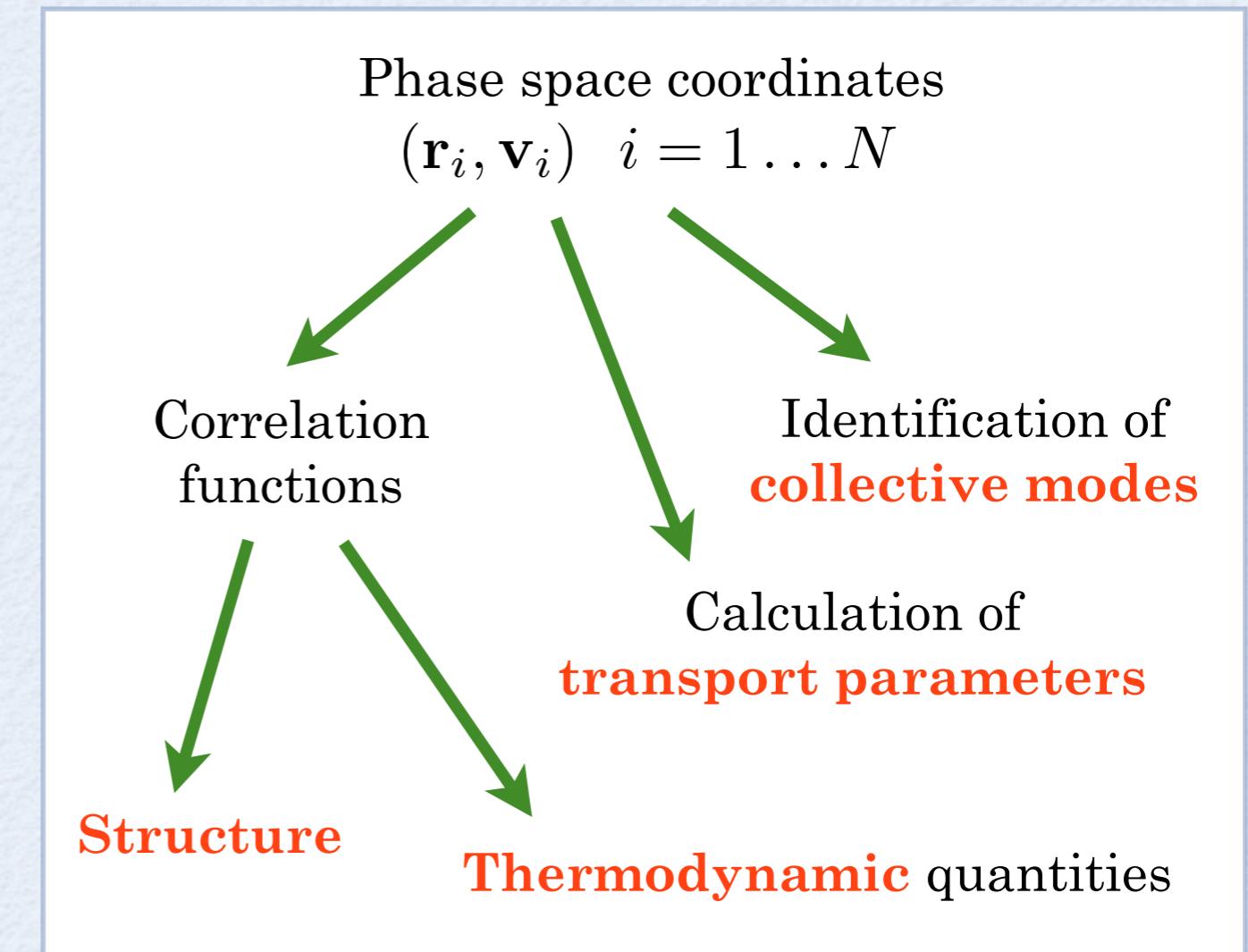
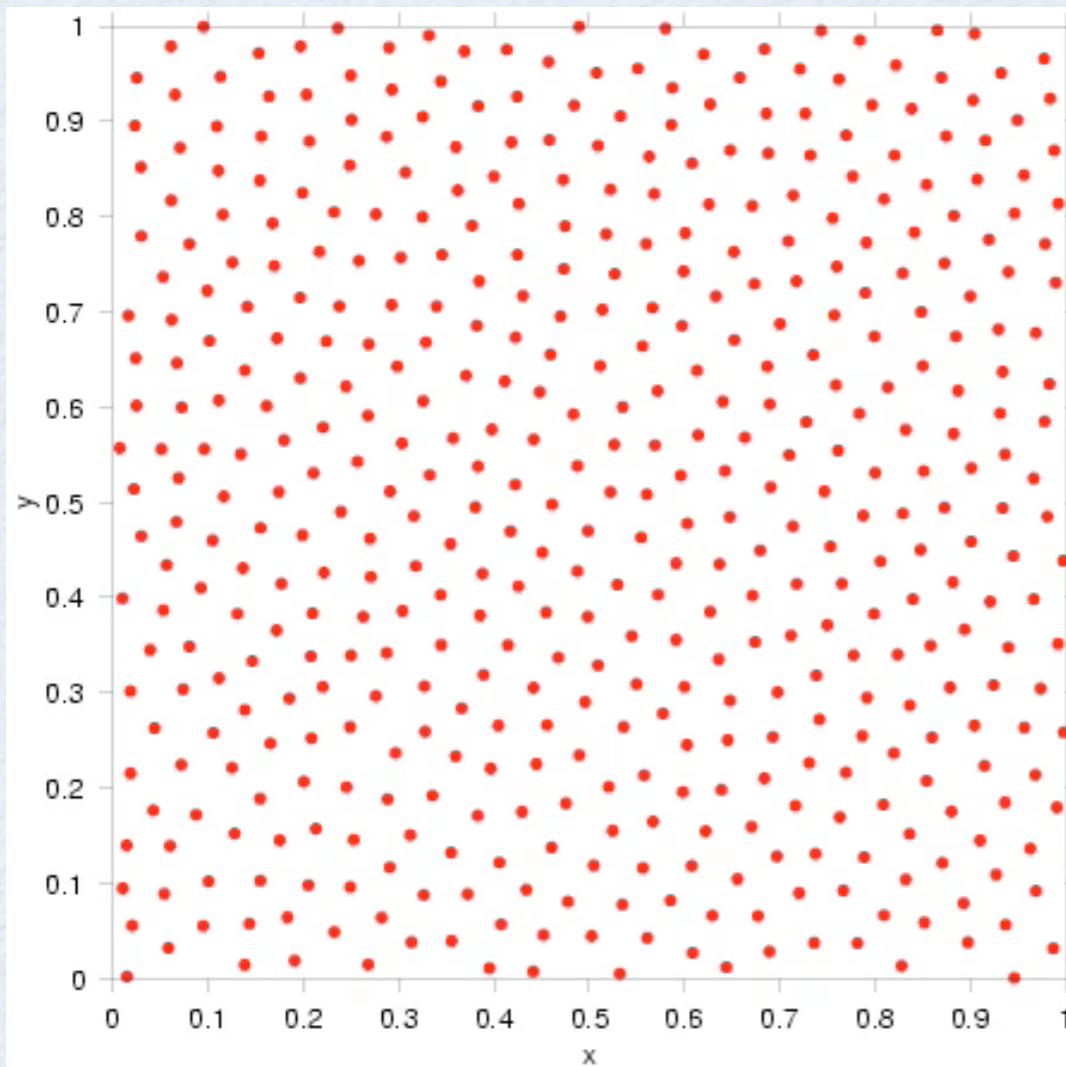


$\Gamma=5, \kappa=1$

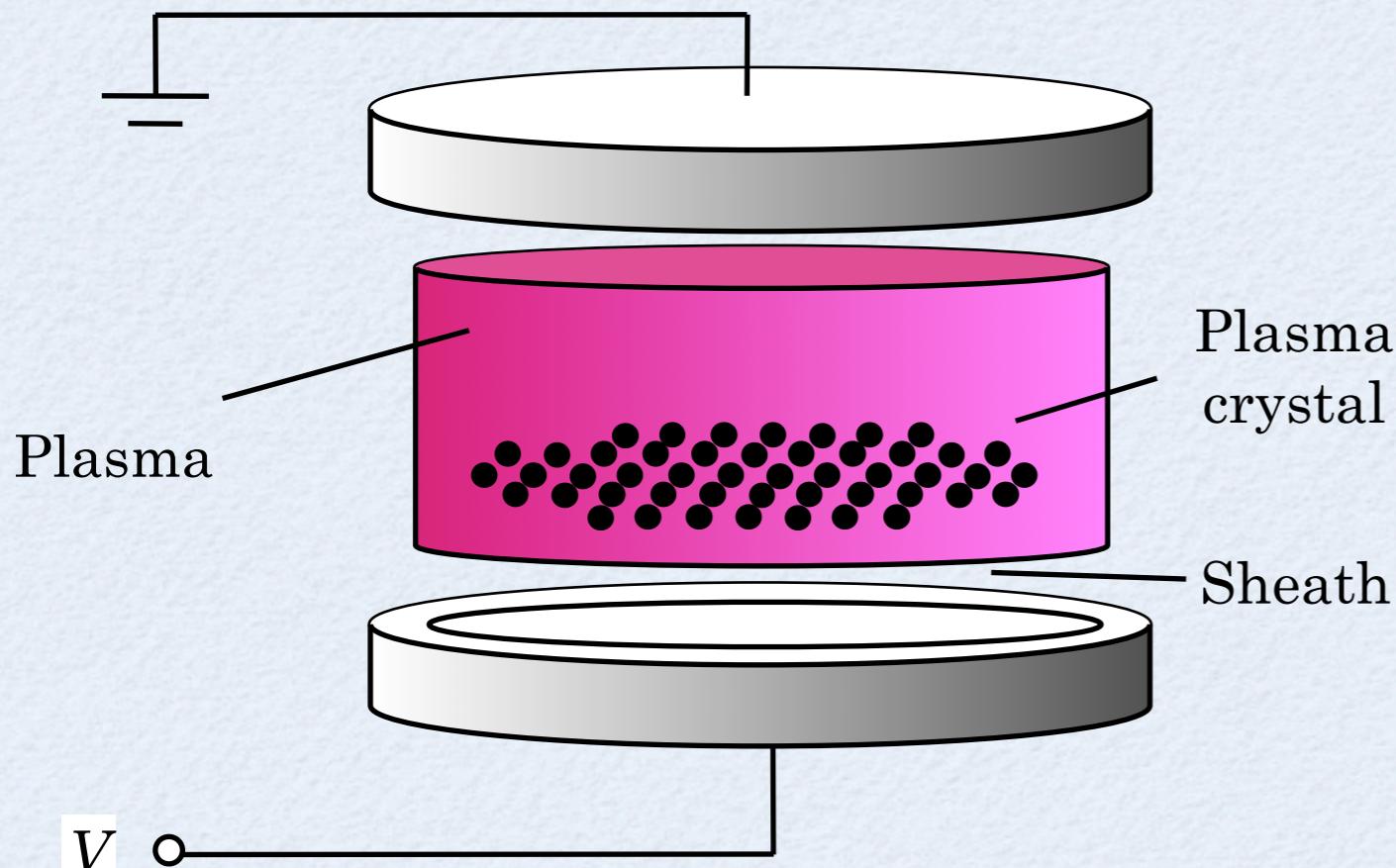


2D frictionless Yukawa liquids

Molecular Dynamics : What do we learn?



It's real: experimental realization of 2D dusty plasma

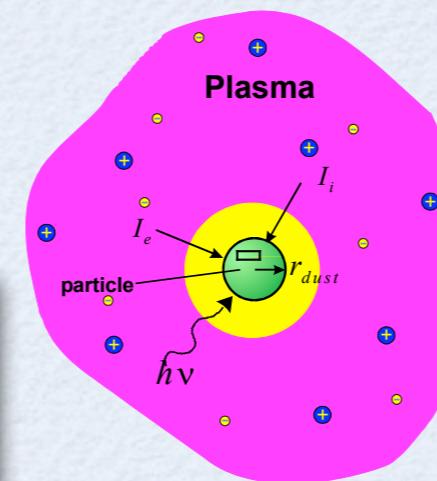
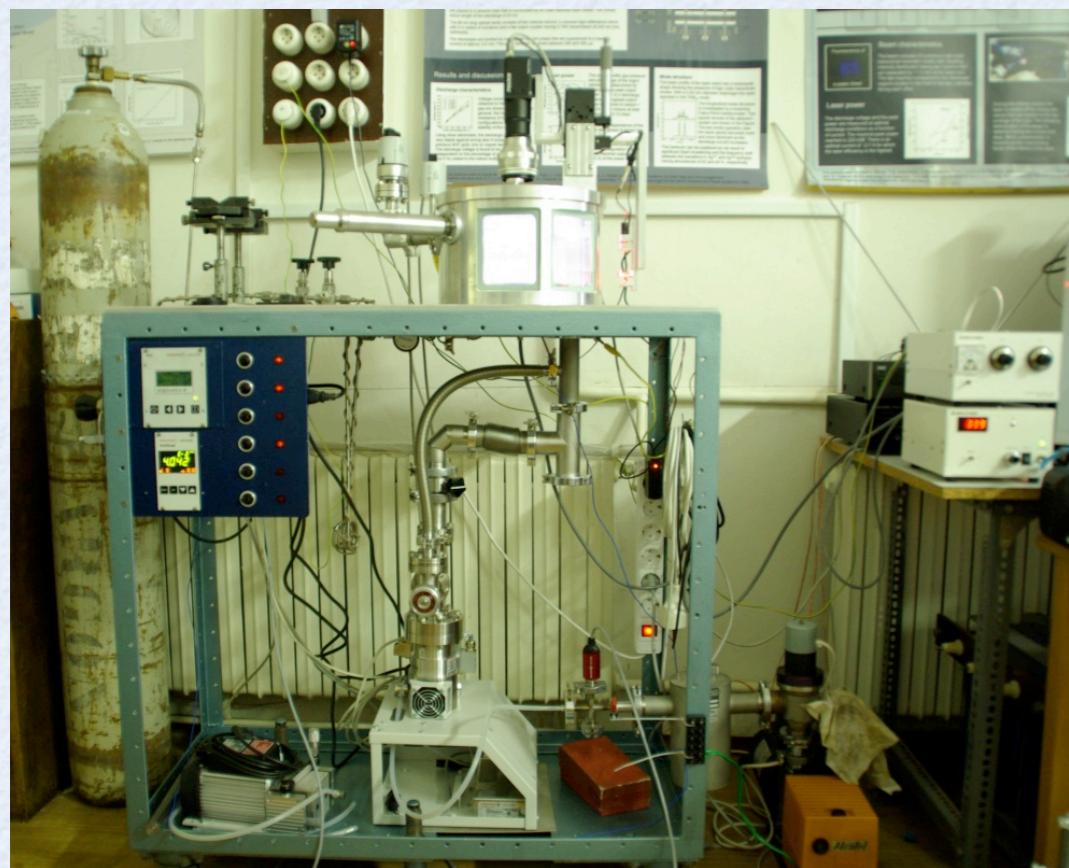


$$\Phi(r) = \frac{1}{4\pi\varepsilon_0} \frac{Q \exp(-r/\lambda_D)}{r} , \quad \kappa = \frac{a}{\lambda_D}$$

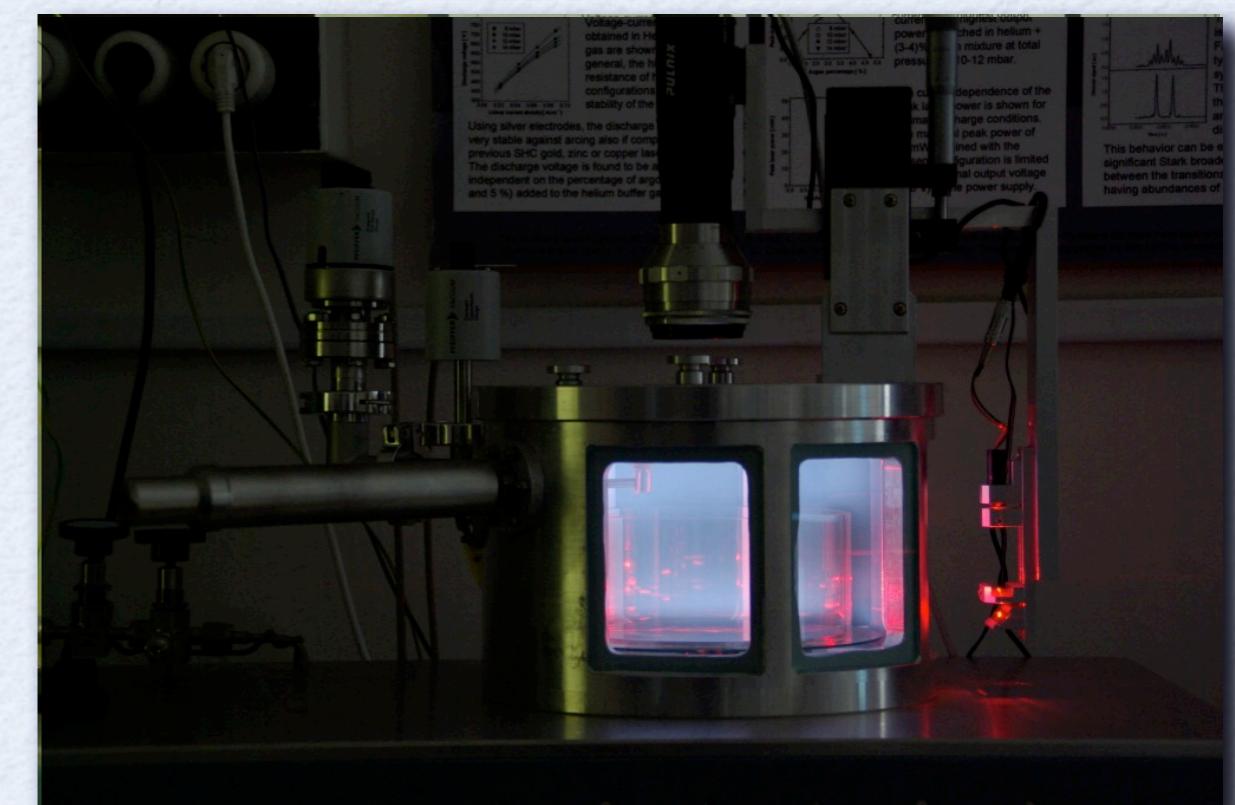
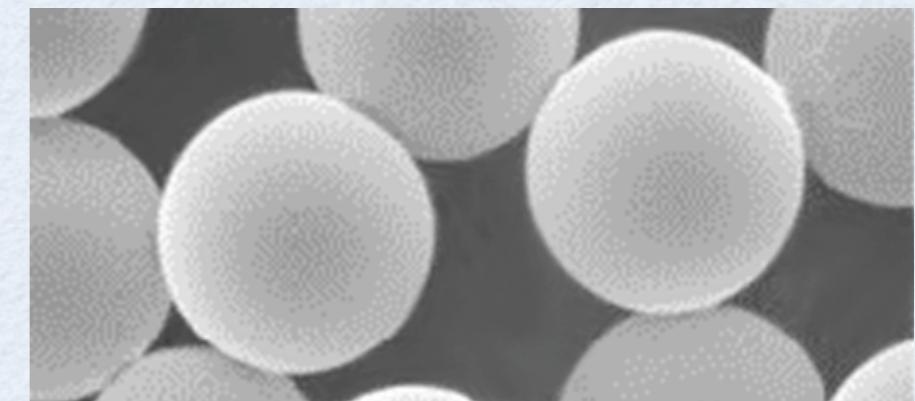
- ❖ Dust particles dispersed in a glow discharge plasma acquire a charge of $\sim 10^4 q_e$
- ❖ Dust layer is levitated due to the balance between electrostatic force and gravity
- ❖ Interaction: screened Coulomb (Yukawa) potential
- ❖ Crystallization at high Γ
- ❖ Quasi-2D confinement
- ❖ Extensive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

Experimental realization of 2D dusty plasma

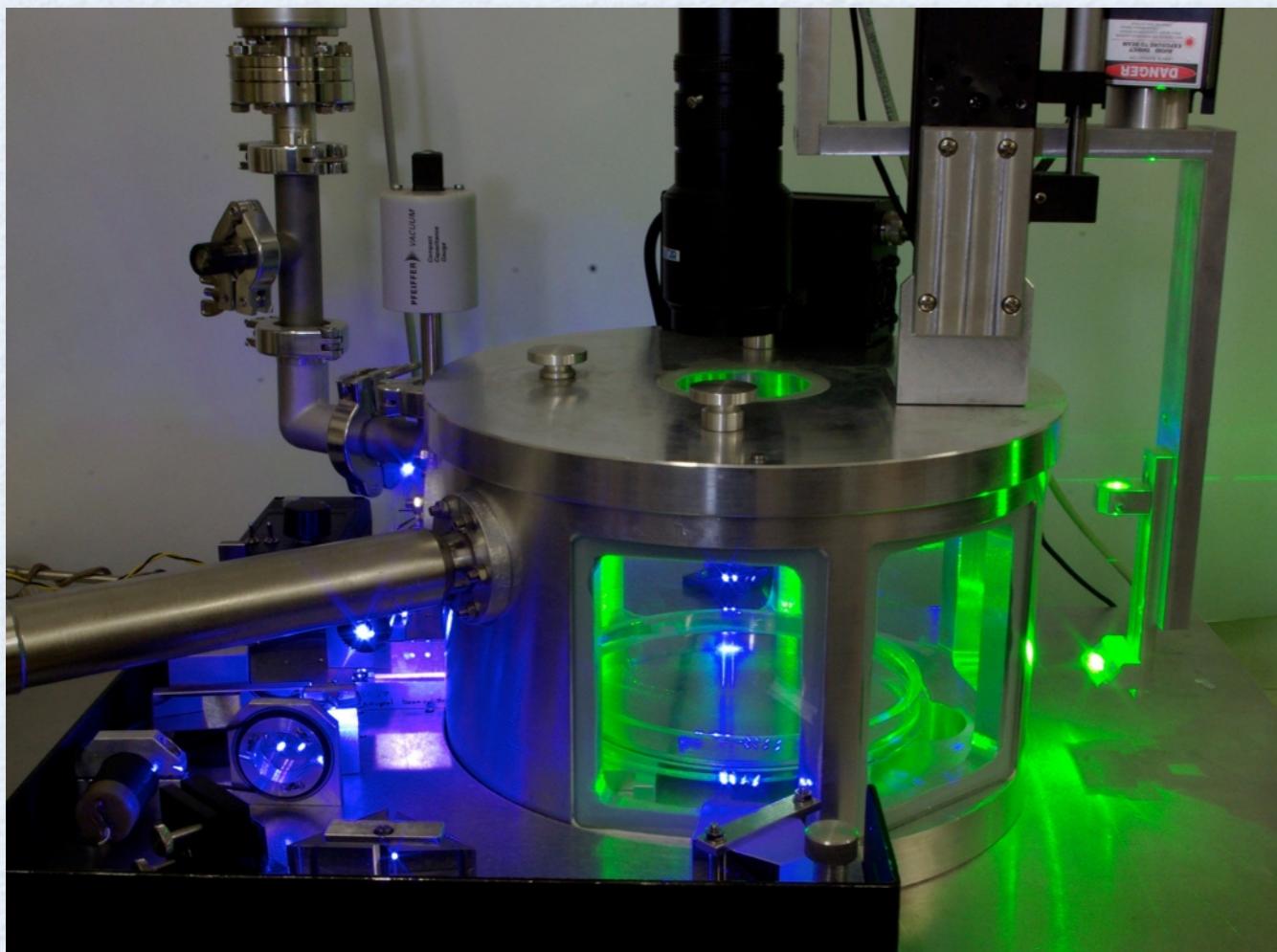
Dusty plasma experiment in
RISSP, Budapest (P. Hartmann)



melamine-formaldehyde microspheres

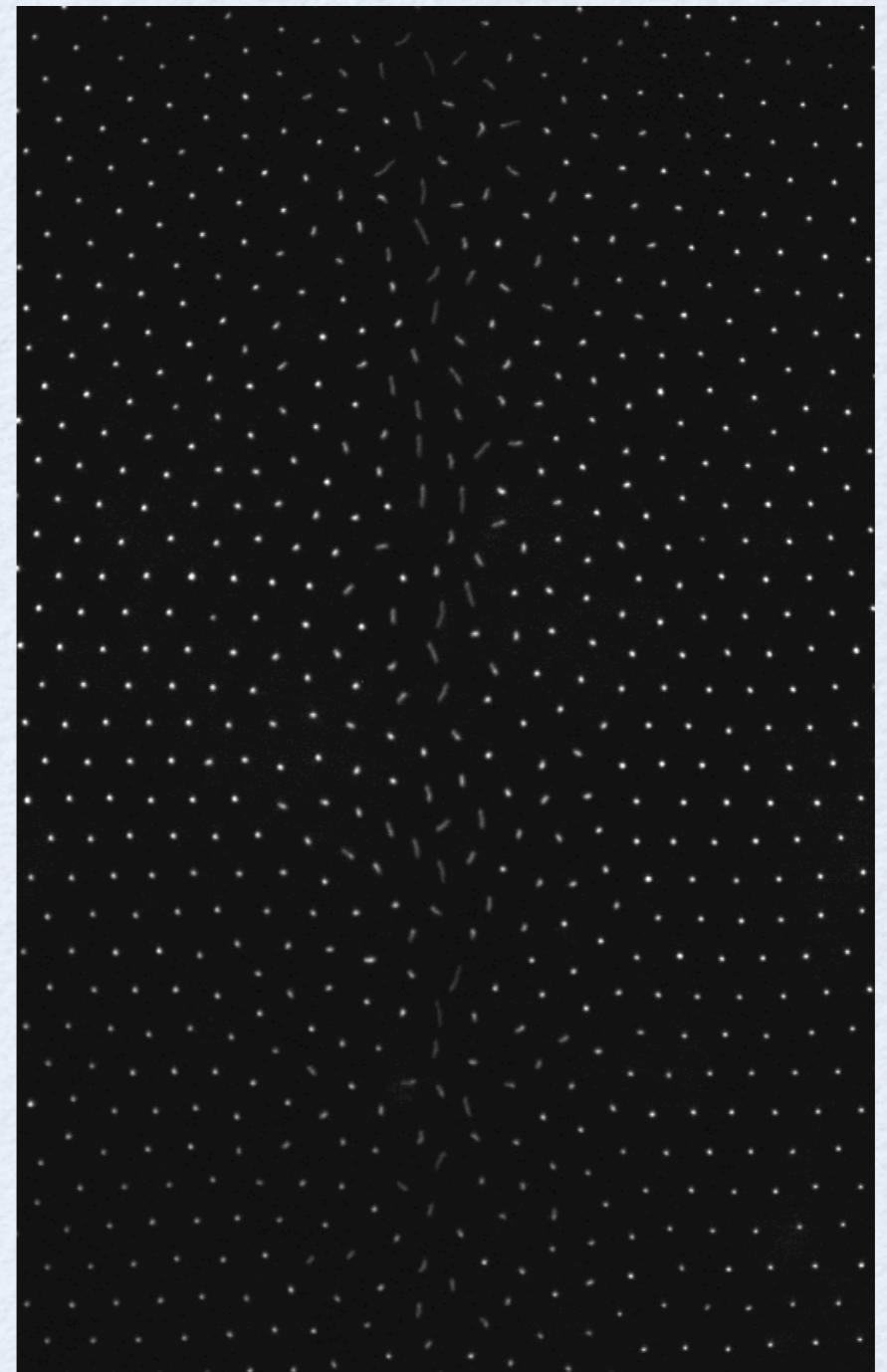


Experimental realization of 2D dusty plasma



Laser manipulation to measure
complex viscosity

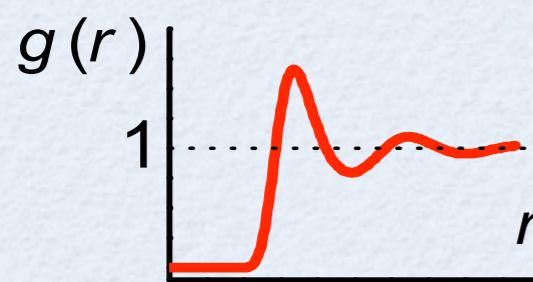
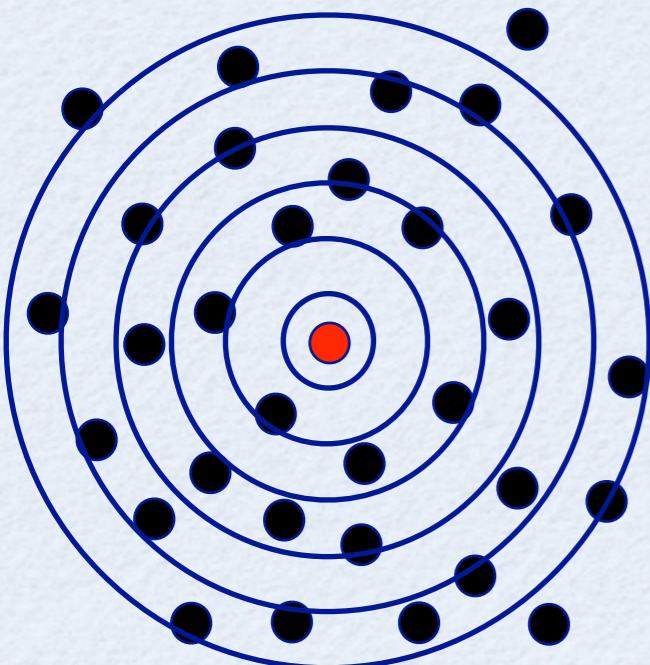
P. Hartmann, M. Cs. Sándor, A.-Zs. Kovács, Z. Donkó:
Phys. Rev. E 84, 016404 (2011).



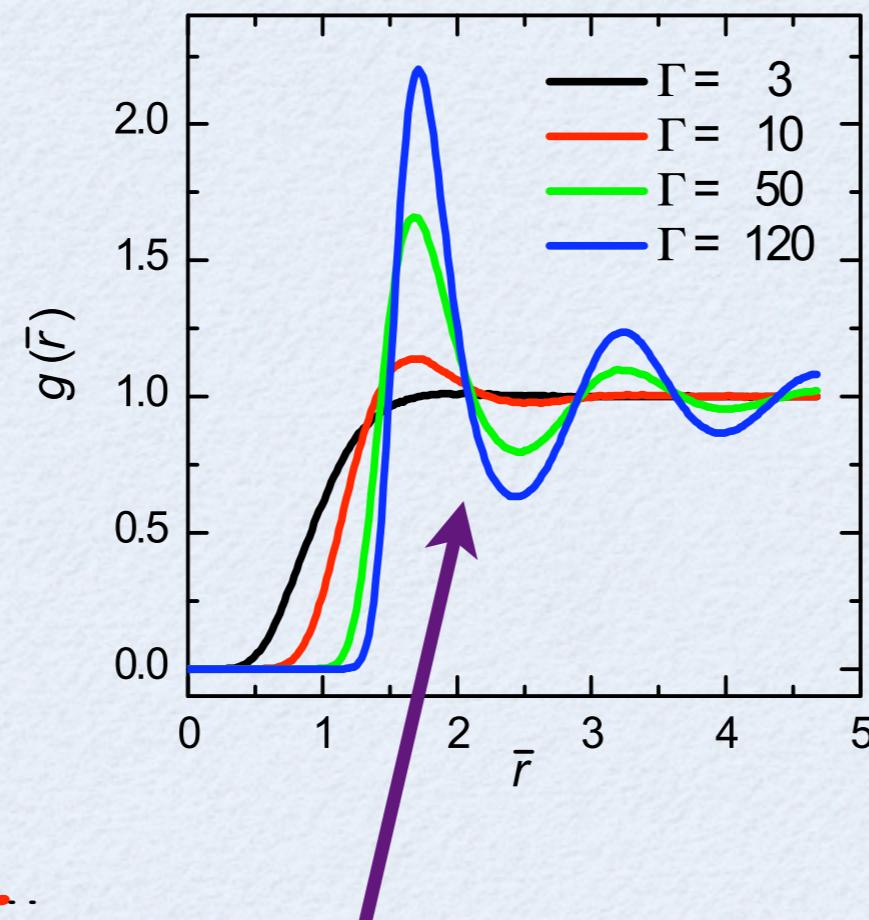
Structural and thermodynamic properties

Pair correlation & thermodynamic properties

Pair correlation function



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



**Strong correlation,
liquid-like structure
at high coupling**

- 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:

$$p = n k_B T - \frac{n^2}{6} \int_0^\infty \frac{\partial \varphi(r)}{\partial r} g(r) 4\pi r^3 dr$$

- Isothermal compressibility:

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

Phase transitions: 3D Coulomb / Yukawa systems

Coulomb (Monte Carlo)

S. G. Brush, H. L. Sahlin and E. Teller,
J. Chem. Phys. 45, 2102 (1966).

$\Gamma \approx 125$

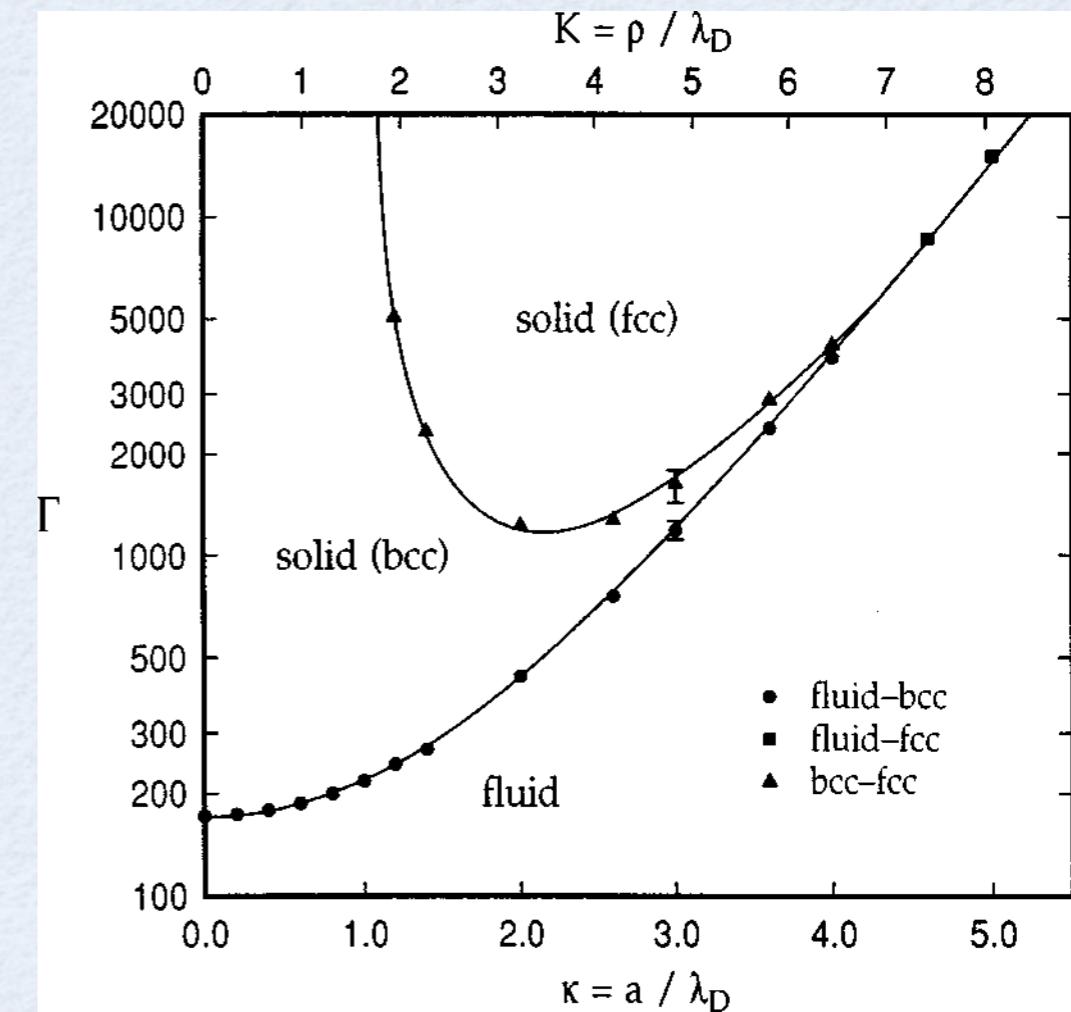
E. L. Pollock and J. P. Hansen
Phys. Rev. A 8, 3110 (1973)

G. S. Stringfellow, H. E. DeWitt and W. L. Slattery,
Phys. Rev. A 41, 1105 (1990).

$\Gamma \approx 175$

Yukawa

S. Hamaguchi, R.T. Farouki and D.H.E. Dubin,
Phys. Rev. E 56, 4671 (1997).



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

$$D = \frac{1}{2} \int_0^\infty C_v dt \quad C_v \equiv \langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle \quad \text{VACF}$$

$$\eta = \frac{1}{V k T} \int_0^\infty C_\eta dt \quad C_\eta \equiv \langle P_{xy}(t) P_{xy}(0) \rangle \quad \text{SACF}$$

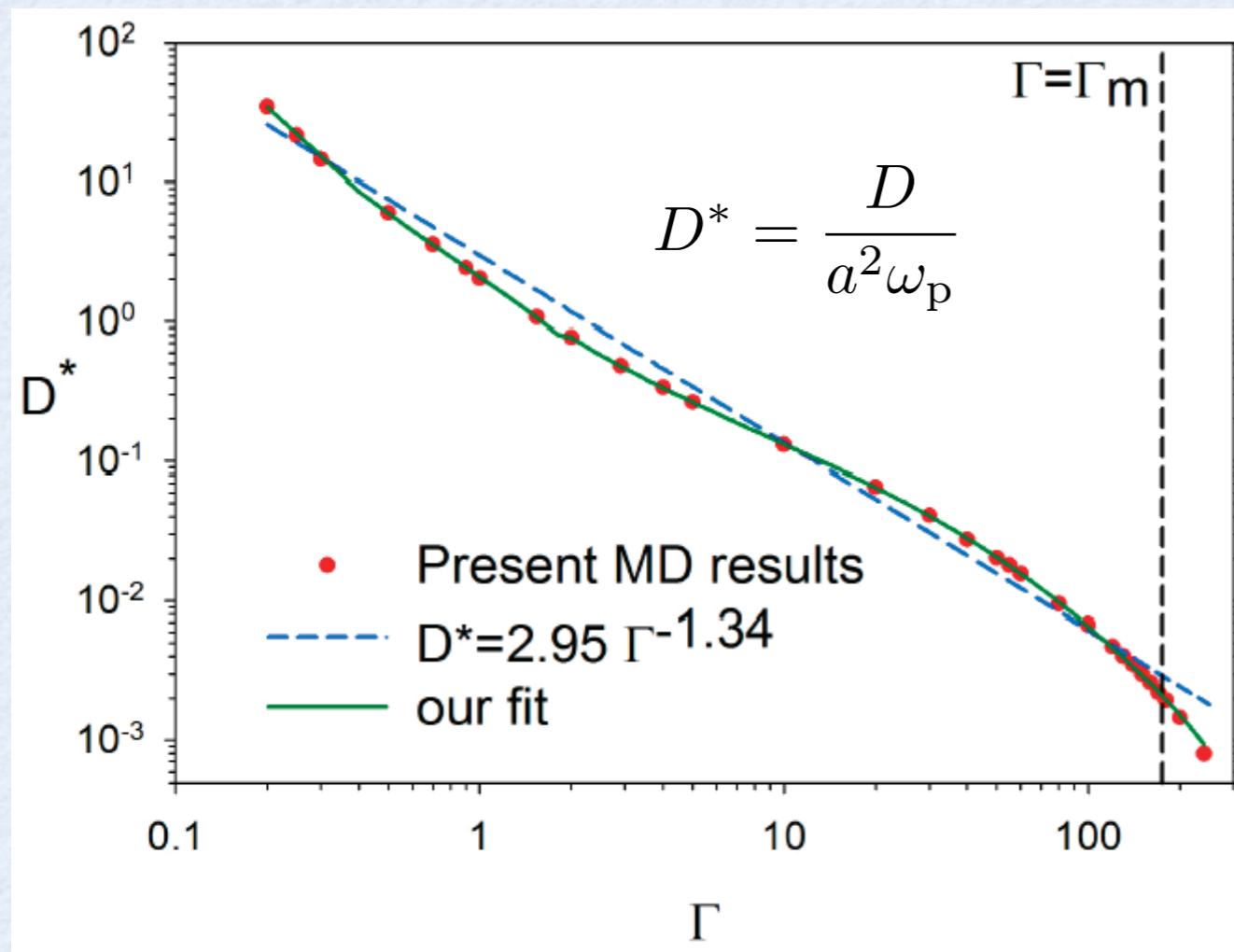
$$\lambda = \frac{1}{V k T^2} \int_0^\infty C_\lambda dt \quad C_\lambda \equiv \langle J_{Qx}(t) J_{Qx}(0) \rangle \quad \text{EACF}$$

Non-Equilibrium Molecular Dynamics:

Perturb the system and measure the response

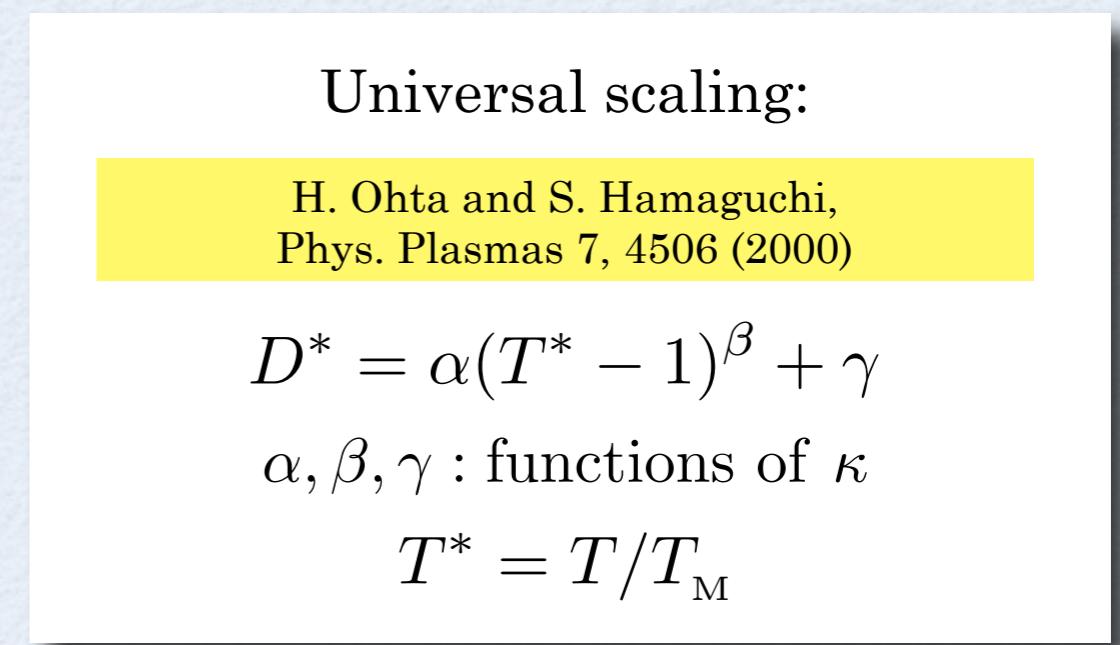
Diffusion coefficient

Coulomb



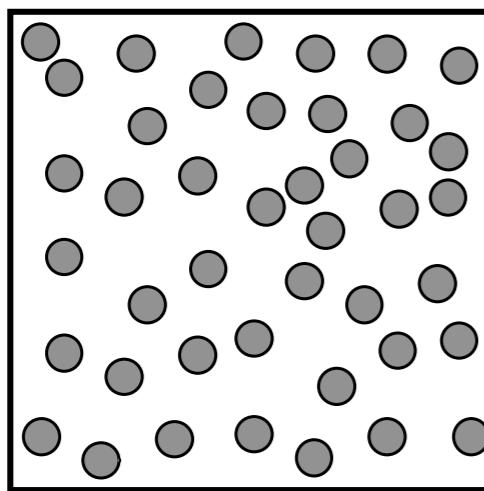
J. Daligault, Phys. Rev. Lett. 96, 065003 (2006)

Yukawa



Shear viscosity: methods (1)

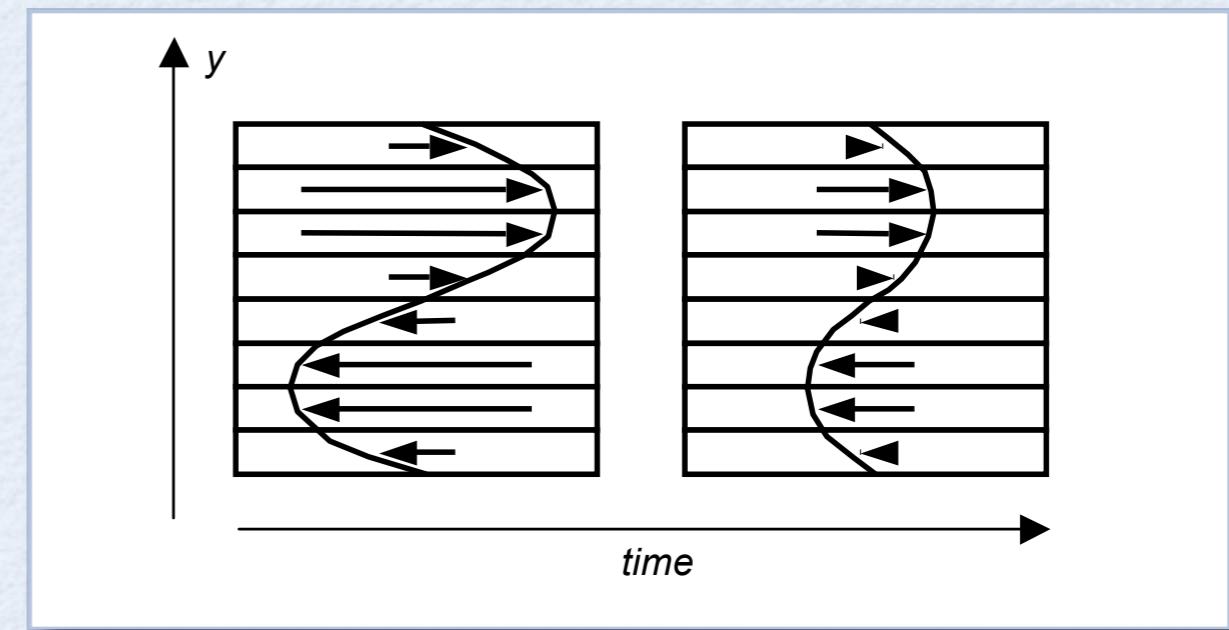
Equilibrium MD



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

$$\eta = \frac{1}{V k T} \int_0^\infty \langle P^{xy}(t) P^{xy}(0) \rangle dt$$

Nonequilibrium (transient perturbation) MD



$$W(y_k) = W_{M0} \sin\left(\frac{2\pi y_k}{L}\right) \quad \frac{\partial v_x}{\partial t} = \frac{\eta}{\rho} \frac{\partial^2 v_x}{\partial y^2}$$

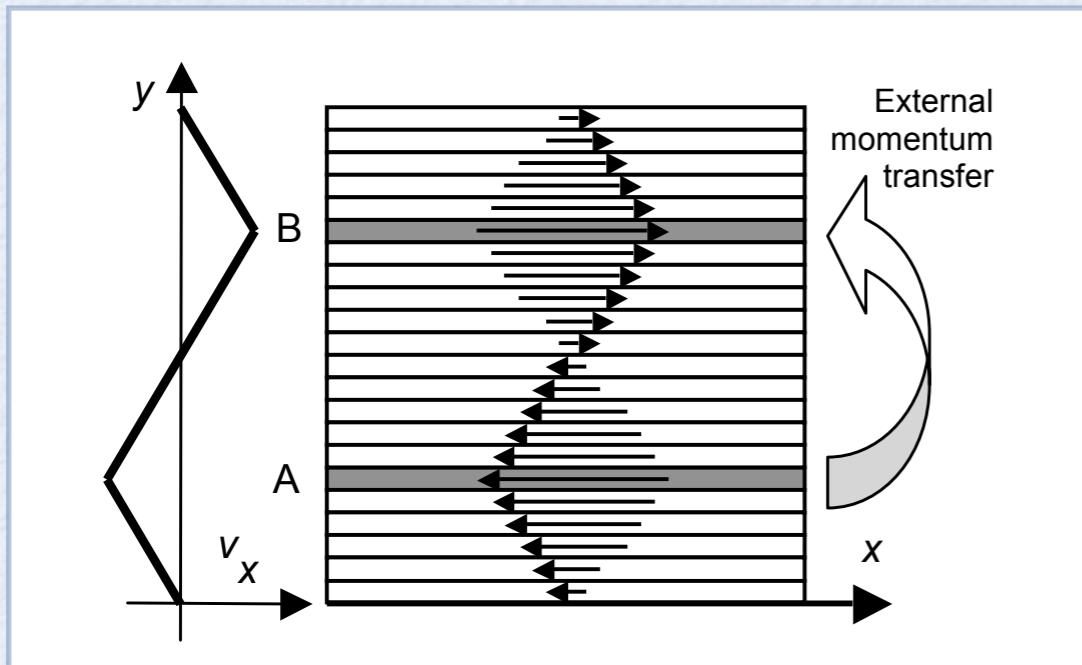
$$W(y, t) = W_{M0} \sin\left(\frac{2\pi y}{L}\right) \exp\left(-\frac{t - t_0}{\tau}\right)$$

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

Z. Donkó and B. Nyíri, Phys. Plasmas 7, 45 2000
 K. Y. Sanbonmatsu and M. S. Murillo, Phys. Rev. Lett. 86, 1215 2001.

Shear viscosity: methods (2)

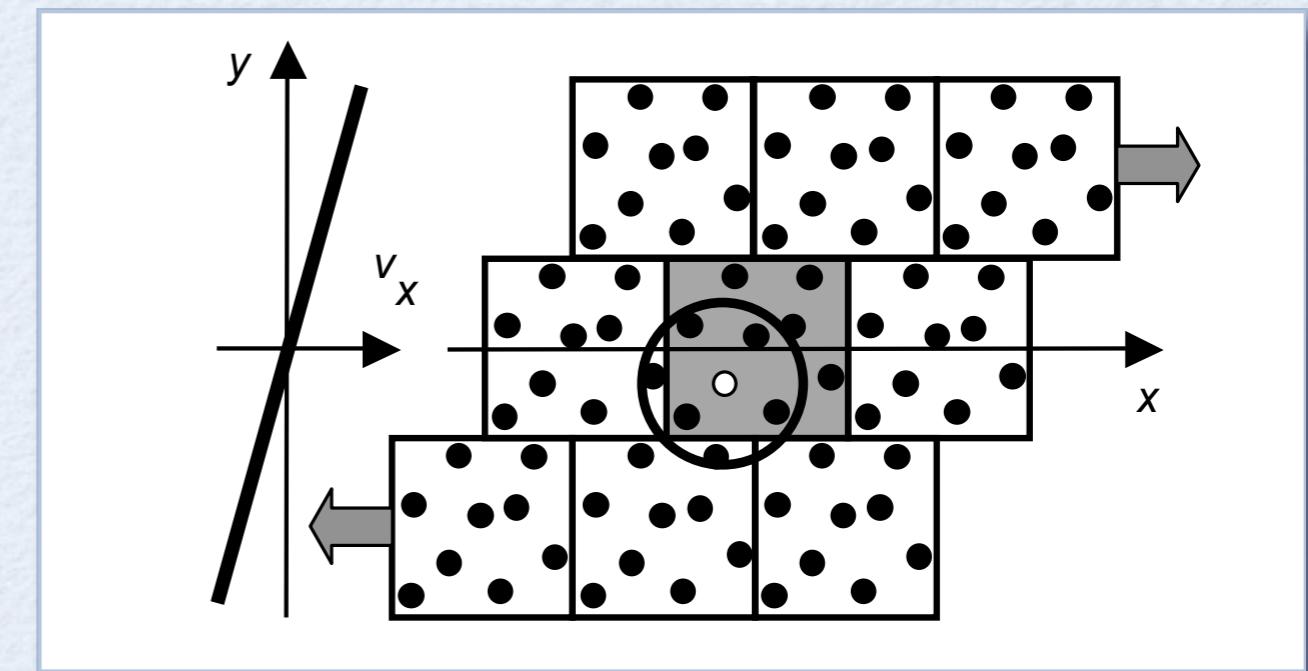
Reverse Molecular Dynamics



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

F. Müller-Plathe,
Phys. Rev. E 59, 4894 (1999).

Homogeneous Shear Algorithm



$$\frac{d\mathbf{r}_i}{dt} = \tilde{\mathbf{p}}_i/m + \gamma y_i \hat{\mathbf{x}} \quad \frac{d\tilde{\mathbf{p}}_i}{dt} = \mathbf{F}_i - \gamma \tilde{p}_{yi} \hat{\mathbf{x}} - \alpha \tilde{\mathbf{p}}_i$$

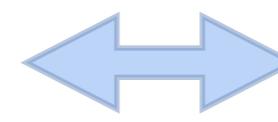
$$\eta = - \lim_{t \rightarrow \infty} \frac{\langle P^{xy}(t) \rangle}{\gamma}$$

D. J. Evans and G. P. Morriss, “Statistical mechanics of nonequilibrium liquids” (Academic Press, 1990)

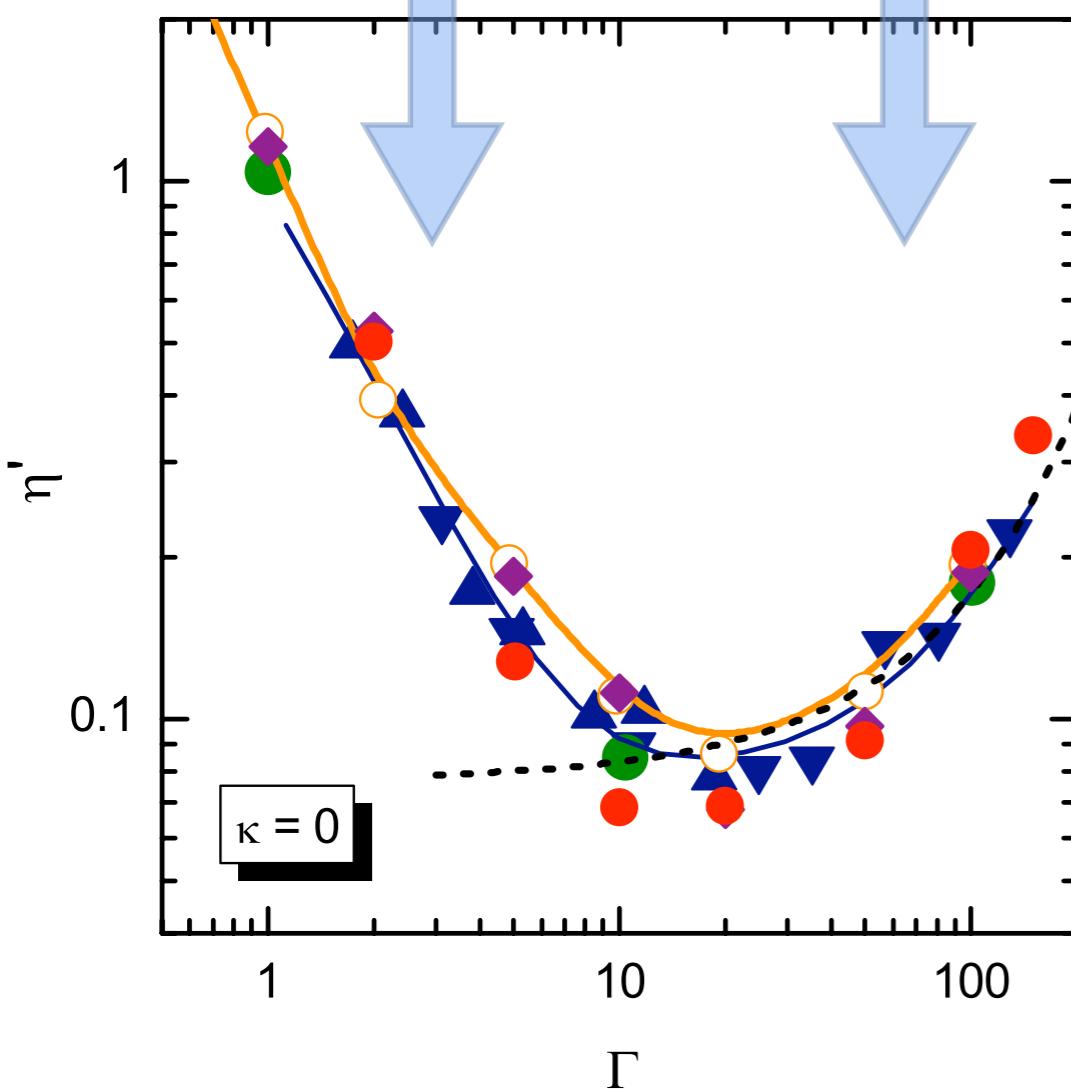
Shear viscosity of 3D Coulomb liquids

Kinetic

Potential



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



Equilibrium MD:

- B. Bernu, P. Vieillefosse, and J. P. Hansen, Phys. Lett. A 63, 301 (1977);
B. Bernu and P. Vieillefosse, Phys. Rev. A 18, 2345 (1978)
- S. Bastea, Phys. Rev. E 71, 056405 (2005)
- - - J. Daligault, Phys. Rev. Lett. 96, 065003 (2006) (Scaled) high Γ 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) ($\kappa = 0.1$)

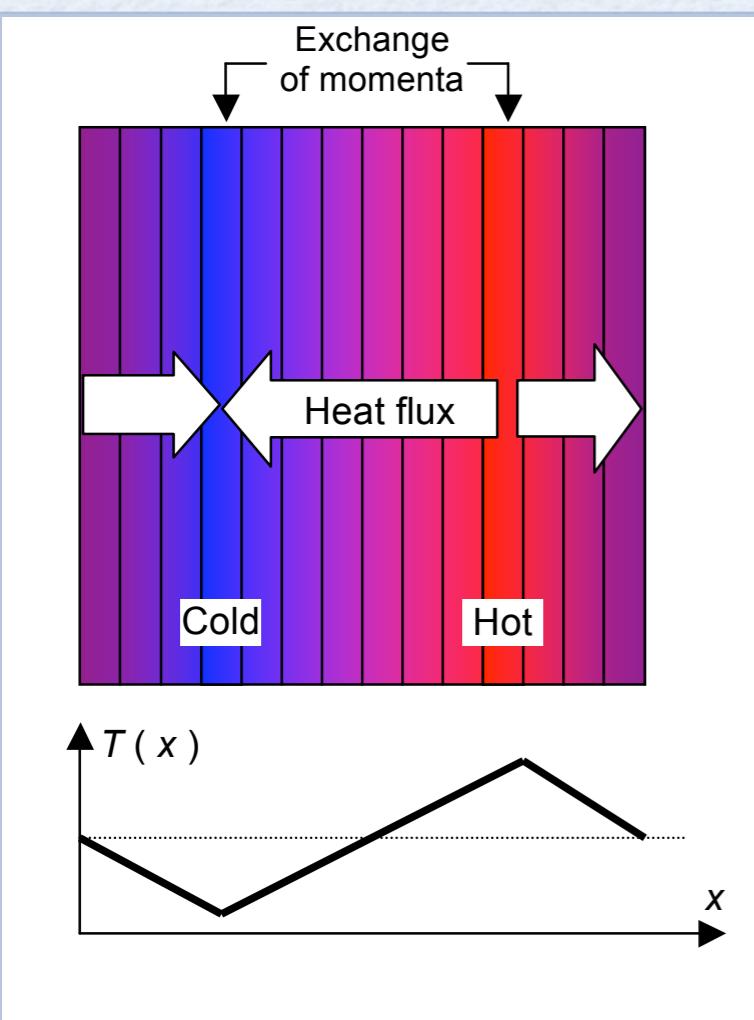
Transient perturbation MD:

- ▲ Donkó & Nyiri ($N = 8192$) Phys. Plasmas 7, 45 (2000) ($N = 8192$)
- ▼ Donkó & Nyiri ($N = 8192$) Phys. Plasmas 7, 45 (2000) ($N = 1024$)

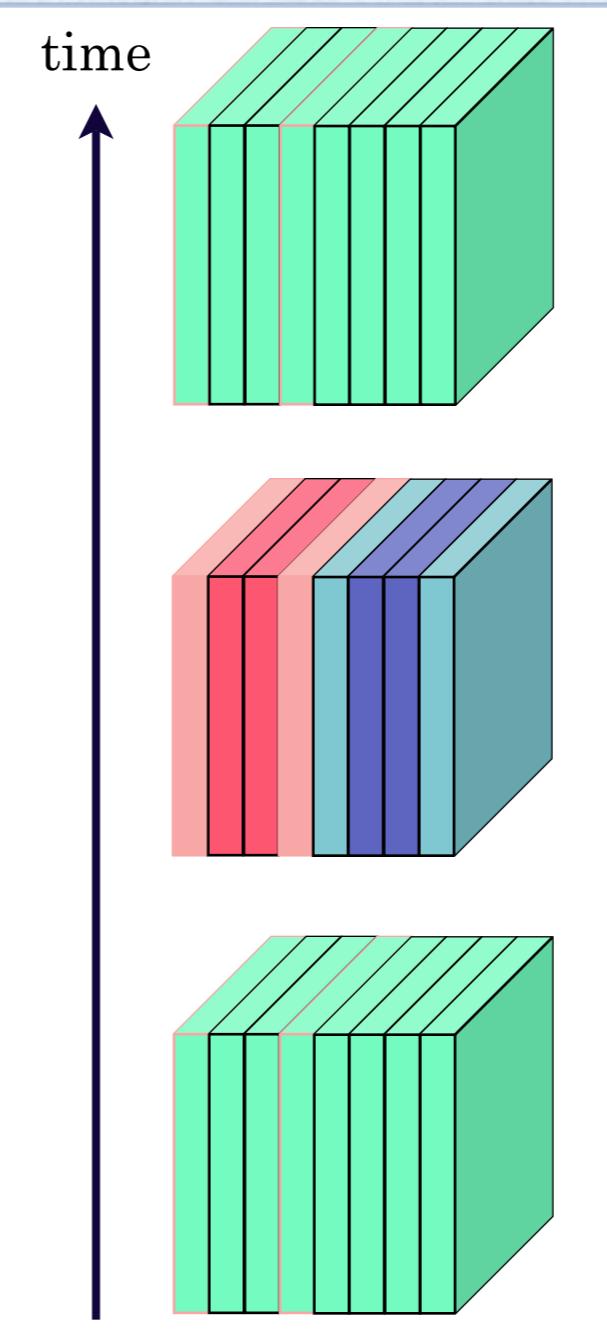
$$\eta' = \frac{\eta}{mn\omega_p a^2}$$

Thermal conductivity: MD methods

Reverse molecular dynamics



F. Müller-Plathe,
J. Chem. Phys. 106, 6082 (1997).



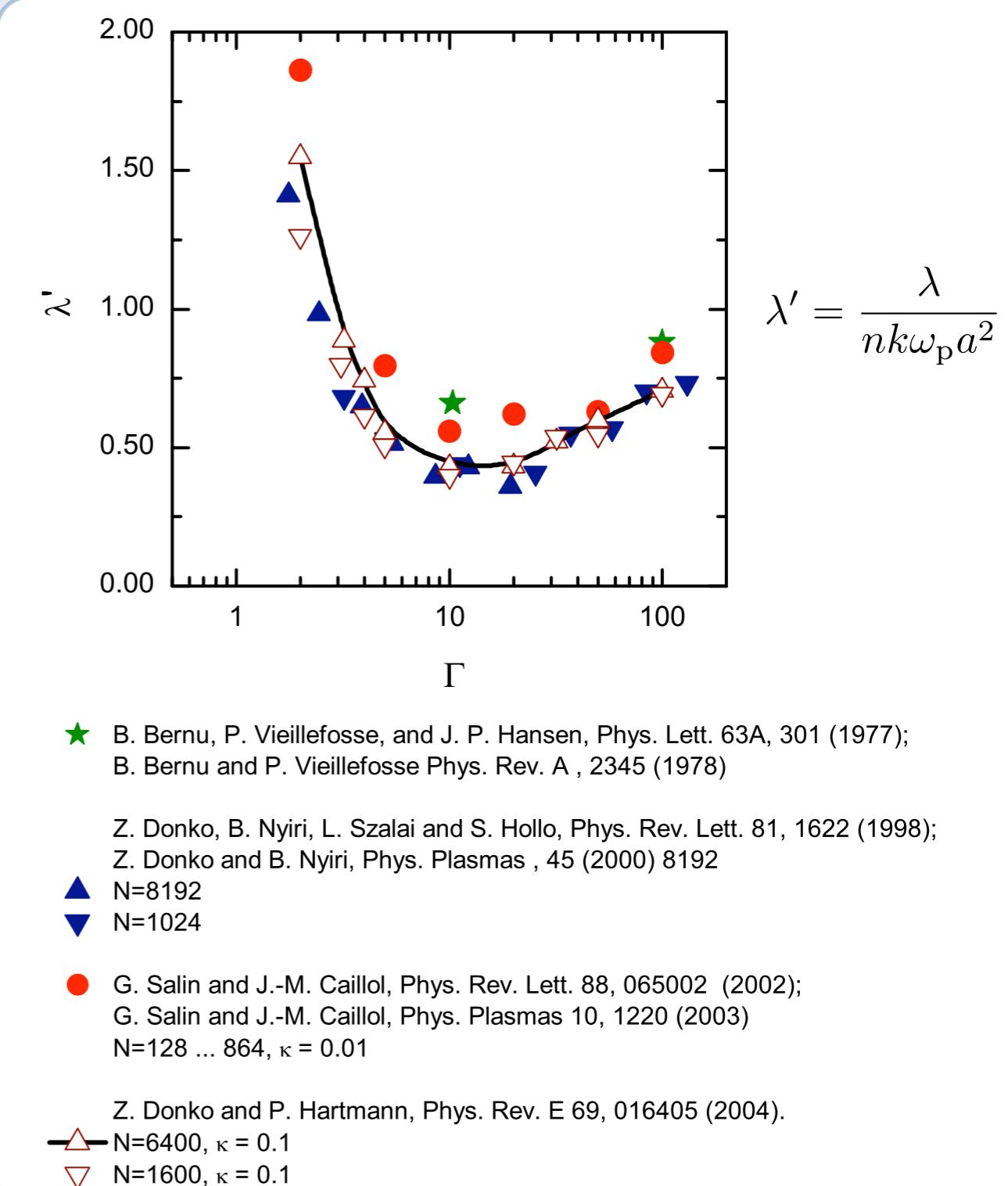
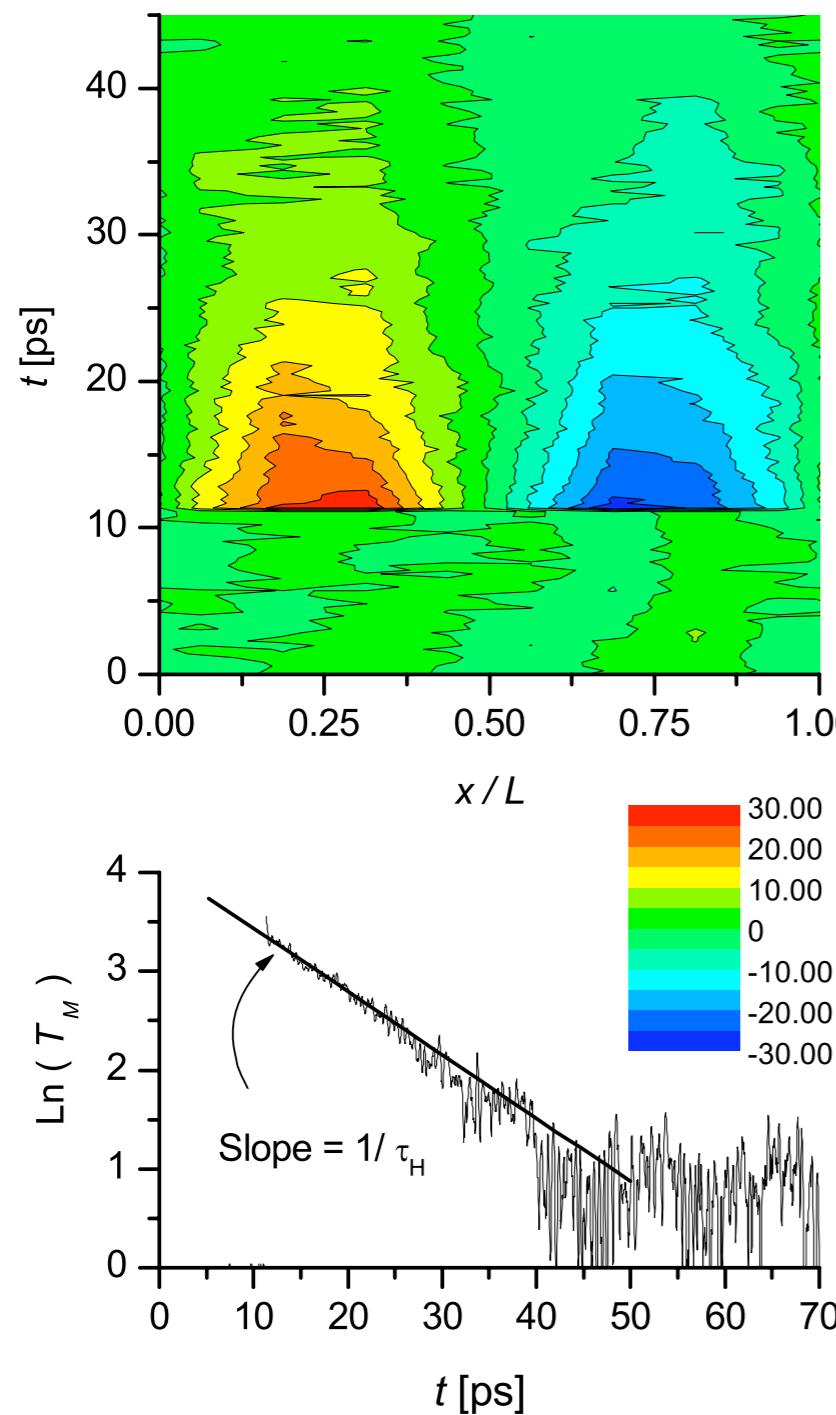
Spatial temperature modulation

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

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

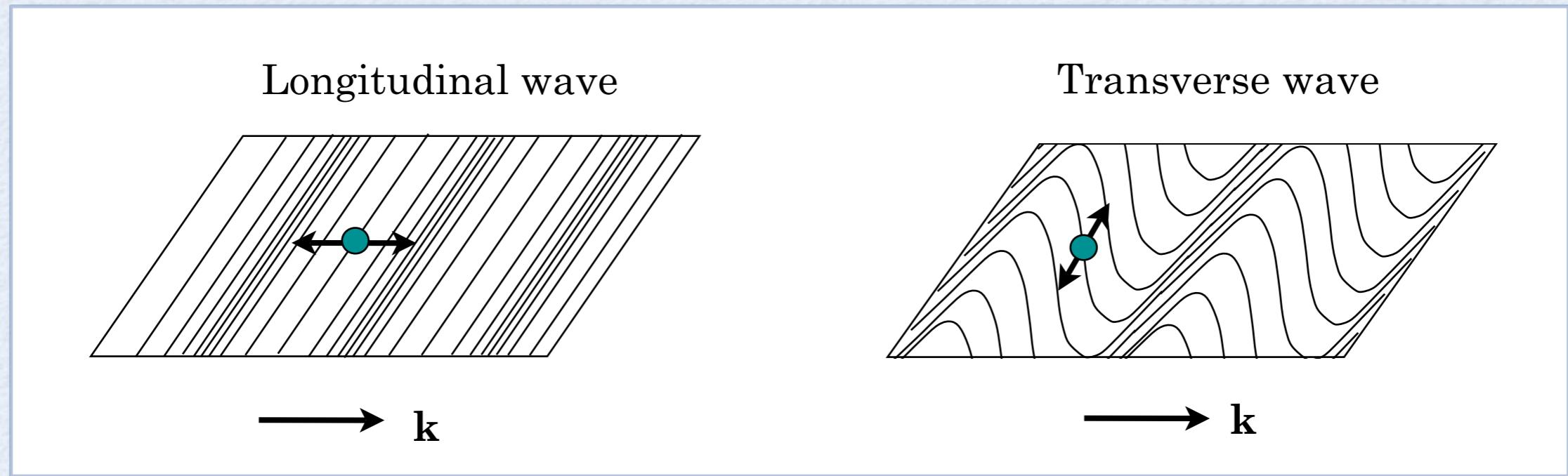
Z. Donkó, B. Nyíri, L. Szalai, and S. Holló,
Phys. Rev. Lett. 81, 1622 (1998).

Thermal conductivity of 3D Coulomb liquids



Collective excitations

Collective excitations in 3D plasma liquids



Microscopic density fluctuations:

$$\rho(k, t) = \sum_{j=1}^N \exp[ikx_j(t)]$$



Dynamical structure function:

$$S(k, \omega) = \frac{1}{2\pi N} \lim_{\Delta T \rightarrow \infty} \frac{1}{\Delta T} |\rho(k, \omega)|^2 \quad \rho(k, \omega) = \mathcal{F}[\rho(k, t)]$$

Microscopic current fluctuations:

$$\lambda(k, t) = \sum_{j=1}^N v_{jx}(t) \exp[ikx_j(t)]$$

$$\tau(k, t) = \sum_{j=1}^N v_{jy}(t) \exp[ikx_j(t)]$$

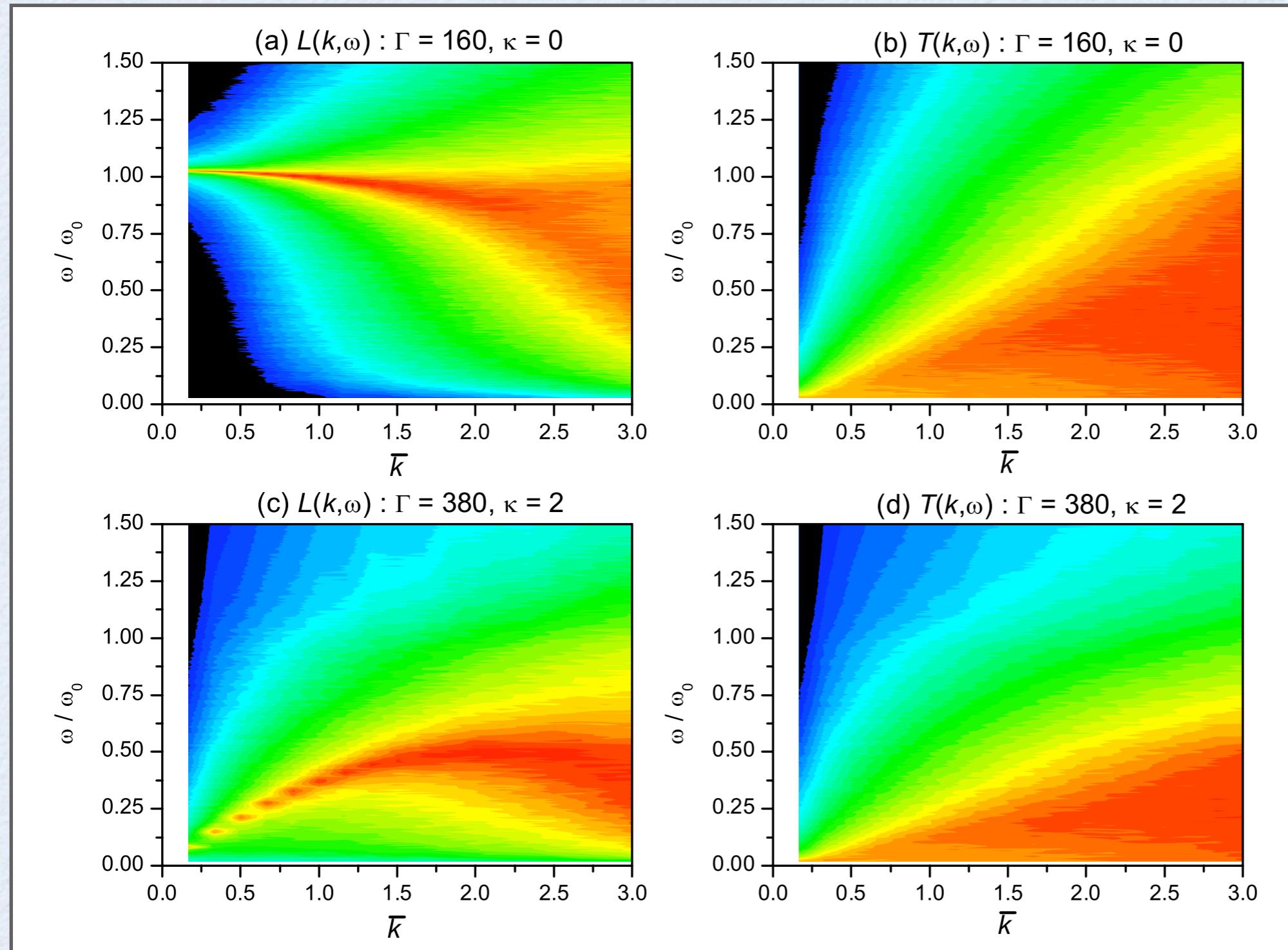


Longitudinal and transverse
current-current fluctuation spectra

$$L(k, \omega)$$

$$T(k, \omega)$$

Collective excitations in 3D plasma liquids



Coulomb:

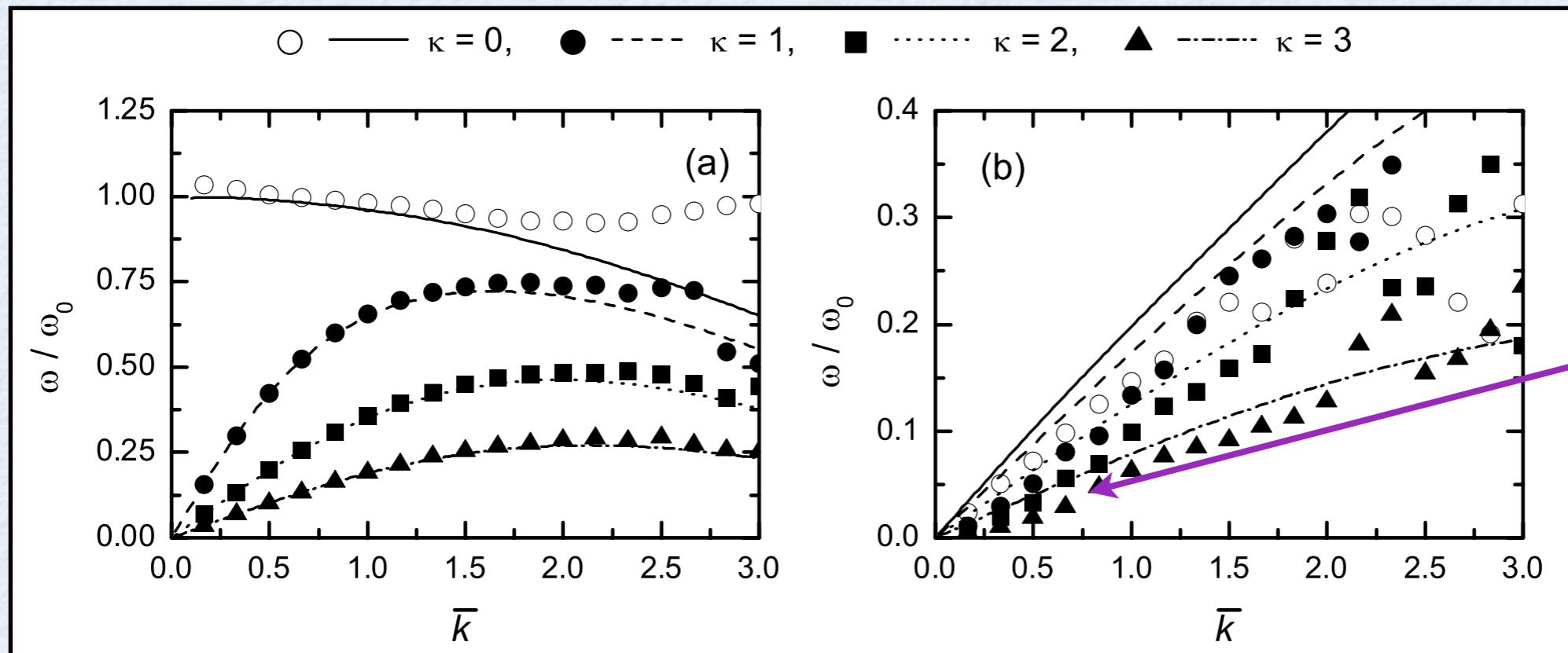
L : const. freq.
T : acoustic

Yukawa:

L : quasi-acoustic
T : acoustic

Collective excitations in 3D liquids: MD vs. theory

QLCA
theory:
↓



$$\Omega_L^2(\mathbf{k}) = \Omega_0^2(\mathbf{k}) + \omega_{0,3D}^2 \frac{\bar{k}^2}{2} \int_0^\infty \Lambda^{3D}(\bar{k}\bar{r}, \kappa\bar{r}) h(\bar{r}) d\bar{r} \quad \Omega_0^2(\mathbf{k}) = \omega_{0,3D}^2 \frac{\bar{k}^2}{\bar{k}^2 + \kappa^2} \quad \text{COMPRESSORIAL MODE}$$

$$\Omega_T^2(\mathbf{k}) = \omega_{0,3D}^2 \frac{\bar{k}^2}{2} \int_0^\infty \Theta^{3D}(\bar{k}\bar{r}, \kappa\bar{r}) h(\bar{r}) d\bar{r} \quad \text{SHEAR MODE}$$

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

$$\Theta^{3D}(x, y) = \frac{1}{2} \left[\frac{e^{-y}}{x} y^2 \left(1 - \frac{\sin(x)}{x} \right) - \Lambda^{3D}(x, y) \right]$$

Z. Donkó, G. J. Kalman & P. Hartmann, J. Phys. Cond. Matter 20, 413101 (2008)

Summary

- 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
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THANK YOU FOR YOUR ATTENTION