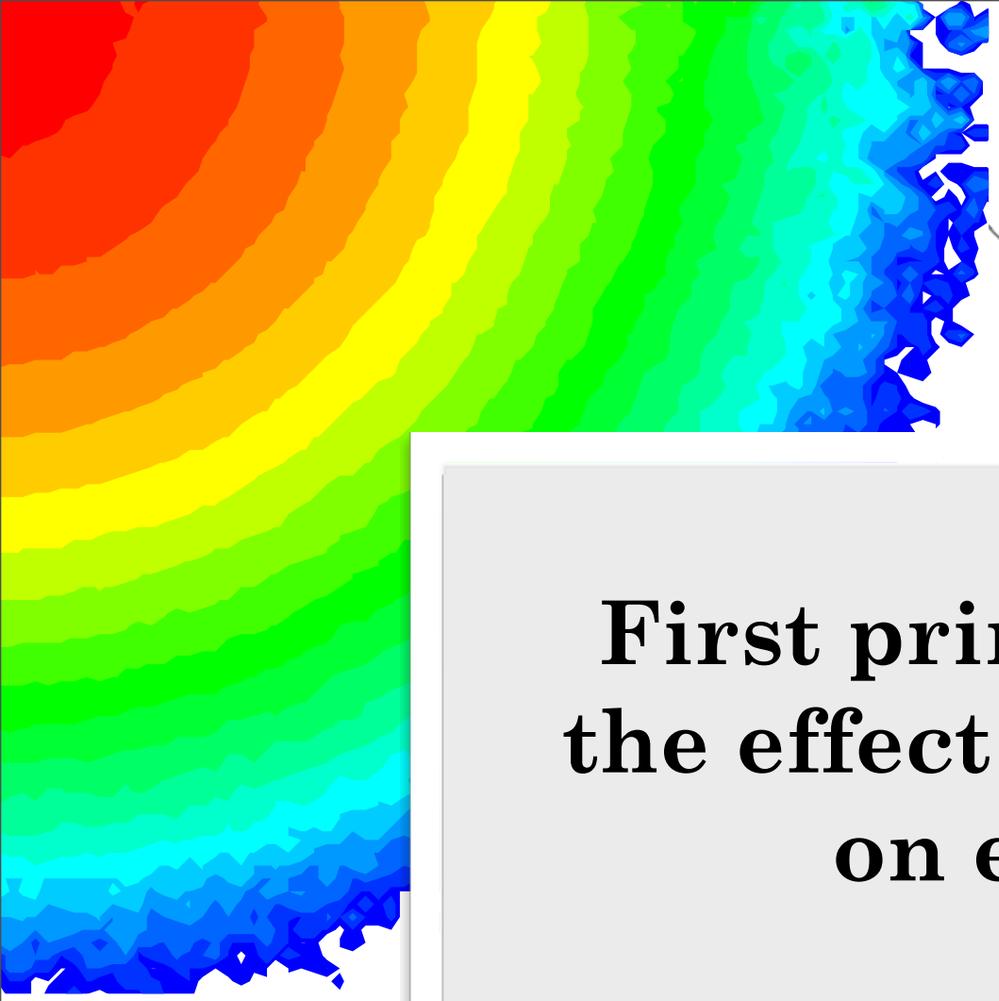

First principles calculation of the effect of Coulomb collisions on electron swarms

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NonEqProc Workshop

SPIG 2014

Introduction / motivation

- **Exact description of electron transport (including boundary effects, relaxation phenomena, non-equilibrium effects, etc.) requires a kinetic approach**. [e.g. R. E. Robson, P. Nicoletopoulos, B. Li, and R. D. White, Plasma Sources Sci. Technol. 17, 024020 (2008); Z. Lj. Petrović, S. Dujko, D. Marić, G. Malović, Ž. Nikitović, O. Šašić, J. Jovanović, V. Stojanović and M. Radmilović-Radenović, J. Phys. D: Appl. Phys. 42, 194002 (2009); S. Dujko, R. D. White, Z. Lj. Petrović, and R. E. Robson, Plasma Sources Sci. Technol. 20, 024013 (2011)]

- In most of the electron swarm studies the effects of **electron-electron (Coulomb) collisions are neglected** due to the very low electron densities assumed

- At elevated ratios of the electron density to the gas number density (ionization degree), $\eta = n_e / n \gtrsim 10^{-6}$, Coulomb collisions, however, **do affect the velocity distribution function** $f(\mathbf{v},\mathbf{r},t)$ of the electrons, as well as their **transport parameters**

Introduction / motivation

- The importance of Coulomb collisions has been pointed out on, e.g.,
  - the **electron temperature in the negative glow** of DC discharges [J. E. Lawler and E. A. Den Hartog, Phys. Rev. A 43, 4427 (1991)]
  - the **trapping of electrons in potential wells** related to striations in gas discharges [Y. B. Golubovskii, V. I. Kolobov, and V. O. Nekuchaev, Phys. Plasmas 20, 101602 (2013)]
  - the **velocity distribution function and transport coefficients** of electrons [D. Loffhagen, Plasma Chem. Plasma Processing 25, 519 (2005)]
  - the **development of negative differential conductivity** [N. L. Aleksandrov, N. A. Dyatko, I. V. Kochetov, A. P. Napartovich, and D. Lo, Phys. Rev. E 53, 2730 (1996)]

- **Inclusion of Coulomb collisions in either of the two fundamental approaches of kinetic theory, the Boltzmann equation analysis and the Monte Carlo simulation, represents a significant challenge, due to the long-range nature of the Coulomb potential. A common approximation adopted in several previous studies has treated the multiple electron-electron interactions as a *succession of binary collisions*** [S. D. Rockwood, Phys. Rev. A 8, 2348 (1973); M. Yousfi, G. Zissis, A. Alkaa, and J. J. Damelincourt, Phys. Rev. A 42, 978 (1990); Y. Weng and M. J. Kushner, Phys. Rev. A 42, 6192 (1990); S. Hashiguchi, IEEE Trans. Plasma Sci. 19, 297 (1991)]

Introduction / motivation

BE & MC:

Coulomb collisions as a succession of binary collisions

Introduction / motivation

BE & MC:

Coulomb collisions as a succession of binary collisions

Combine tools of transport theory and many-body physics

Monty Python

Computational methods / Monte Carlo

Monte Carlo transport calculations: **we play the game**

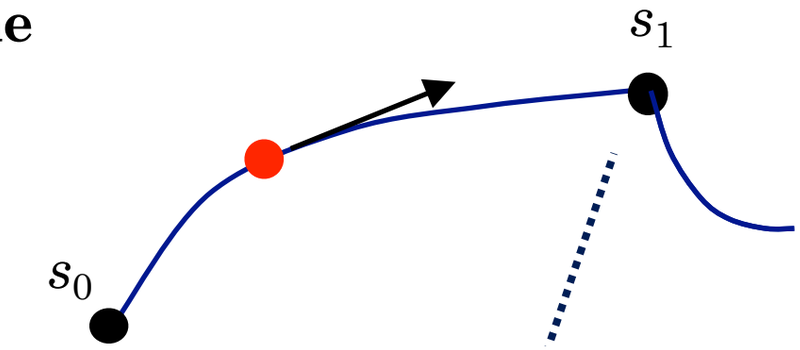
Charged particle - neutral interaction (usually)

1) Equation of motion:

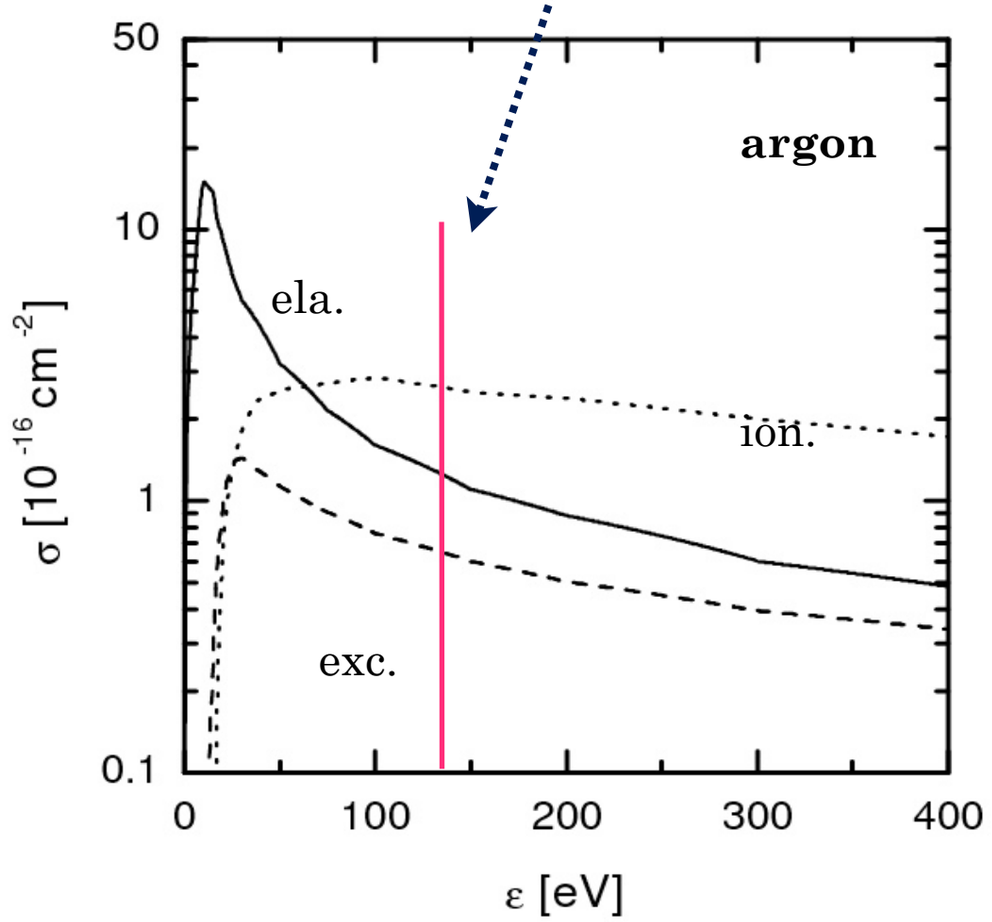
$$m\frac{\mathrm{d}^2\mathbf{r}}{\mathrm{d}t^2} = e\mathbf{E}$$

2) Probability of collision (cold gas):

$$P_{\mathrm{coll}} = 1 - \exp[-n_g\sigma_t(v)v\Delta t]$$



Select type according to the probability $P_k = \dfrac{\sigma_k(\varepsilon)}{\sigma_{\mathrm{T}}(\varepsilon)}$



- Processes
  - elastic scattering,
  - electronic excitation,
  - ionization

- Chosen in a probabilistic manner:
  - free path,
  - type of collision,
  - new direction

Computational methods / MC + MD

Two well-established methods

Monte Carlo transport calculations

Transport theory

Charged particle - neutral interaction (usually)

Interaction of the classical electron gas with the neutral buffer gas
MC

$$P_{\mathrm{coll}} = 1 - \exp[-n_g \sigma_t(v) v \Delta t]$$

Molecular dynamics simulation

Many-body physics

Charged particle - charged particle interaction (usually)

Simultaneous many-body interaction within the classical electron gas
MD

$$m\frac{\mathrm{d}^2\mathbf{r}_i}{\mathrm{d}t^2} = \sum_{i\neq j}\mathbf{F}_{ij} + e\mathbf{E}$$

Completely first principles
No need for binary approximation of electron-electron "collisions"

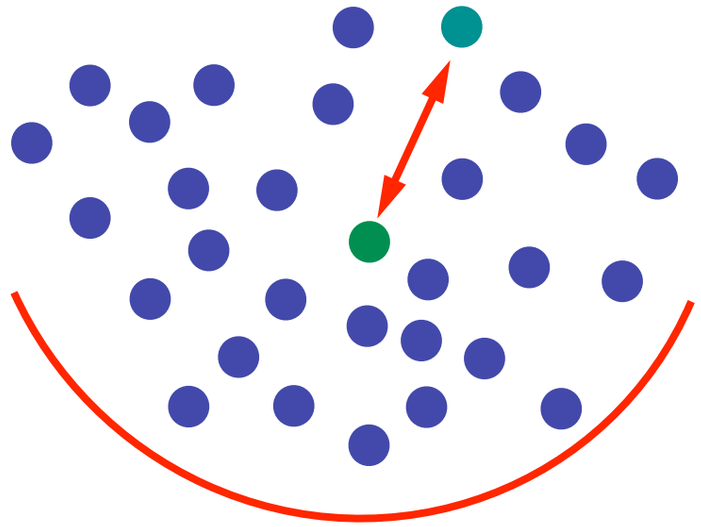
[Z. Donkó, Physics of Plasmas 21, 043504 (2014)]

⇨ How to solve the equation of motion and how to calculate the sum of forces?

Molecular Dynamics (MD) simulation basics

Example: *finite system with external confinement*

- Given N particles (same charge, mass, …)
- Given interaction potential
- Given external potential
- The job is to solve the equation of motion

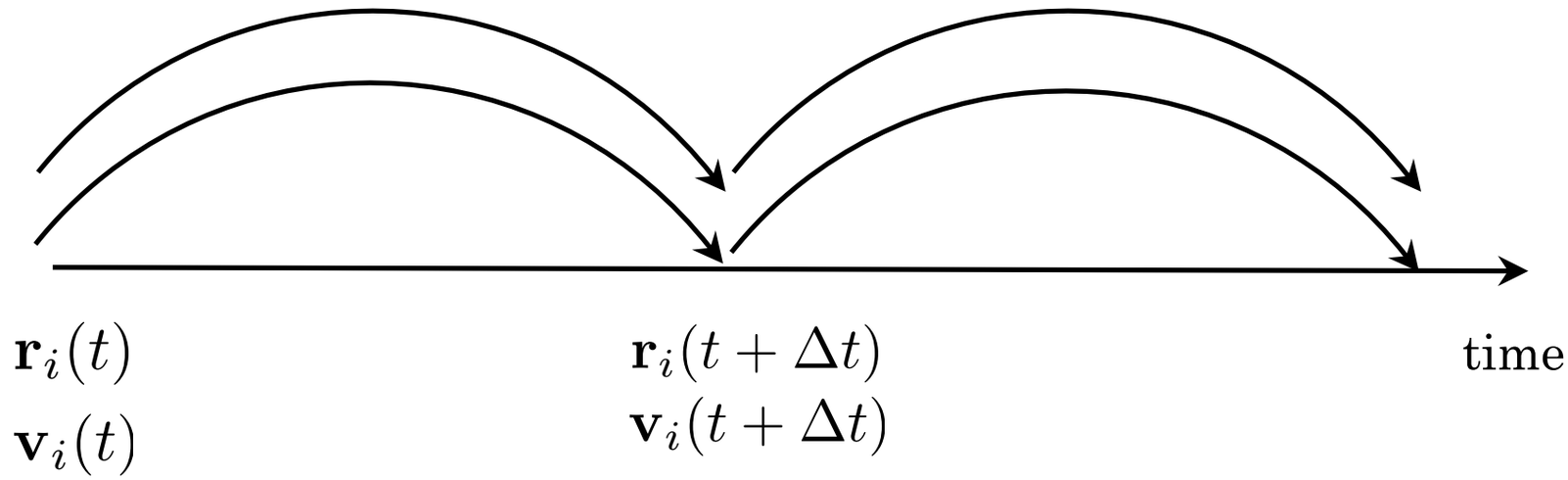


$$\frac{\mathrm{d}^2\mathbf{r}_i(t)}{\mathrm{d}t^2} = \frac{\mathbf{F}_i(t)}{m} \quad \Leftarrow \text{ includes confinement}$$

$$\Downarrow$$

$$\frac{\mathrm{d}\mathbf{r}_i(t)}{\mathrm{d}t} = \mathbf{v}_i(t)$$

$$\frac{\mathrm{d}\mathbf{v}_i(t)}{\mathrm{d}t} = \frac{\mathbf{F}_i(t)}{m} = \mathbf{a}_i(t)$$



Discretization:
"Velocity-Verlet" integration scheme $\Rightarrow$

$$\mathbf{r}_i(t+\Delta t) = \mathbf{r}_i(t) + \mathbf{v}_i(t)\Delta t + \frac{1}{2}\mathbf{a}_i(t)\Delta t^2$$

$$\mathbf{v}_i(t+\Delta t) = \mathbf{v}_i(t) + \frac{\mathbf{a}_i(t) + \mathbf{a}_i(t+\Delta t)}{2}\Delta t$$

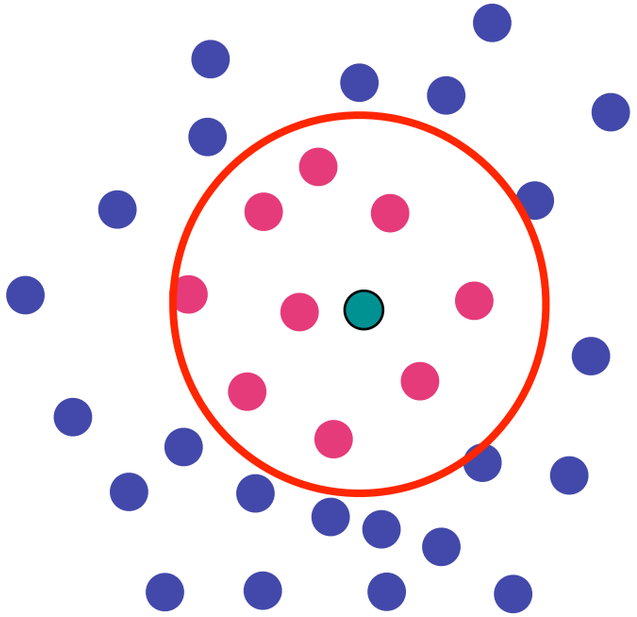
Calculation of the forces / short-range potentials

Short – range interaction potentials

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

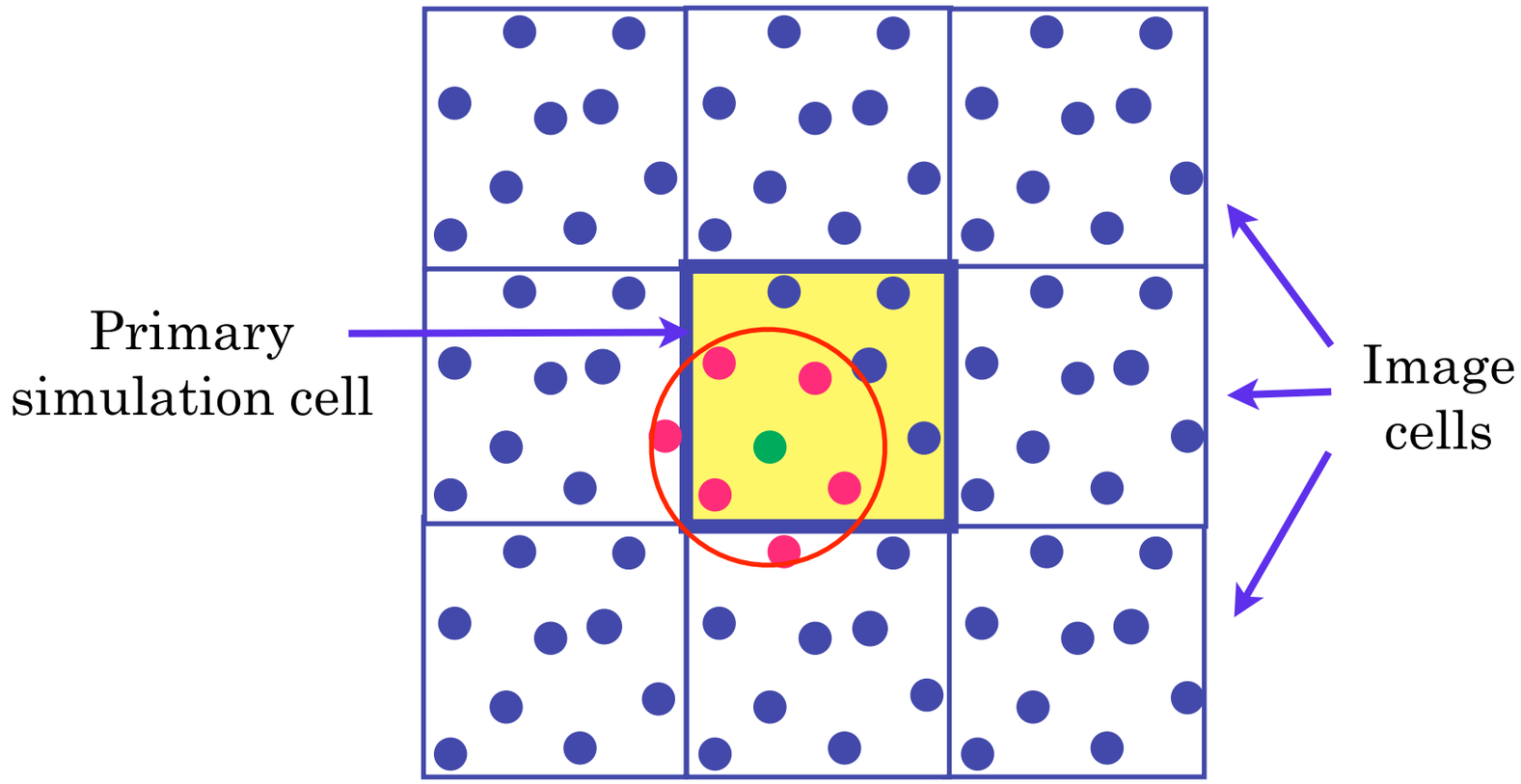
$$\mathbf{F}_i(t) = \sum_{r_{ij}<r_C} \mathbf{F}_{i,j}(t)$$

Finite system

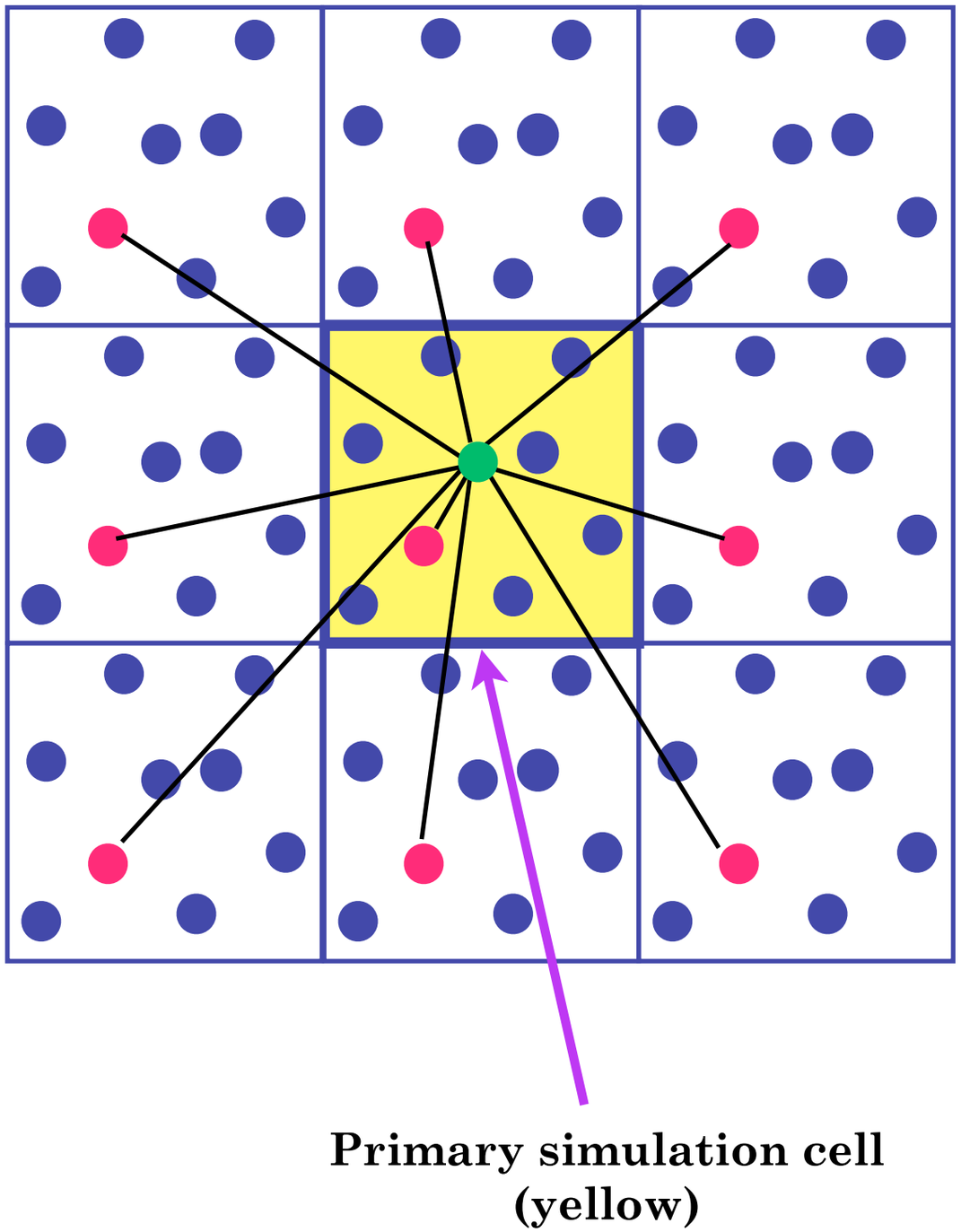


Infinite system

PERIODIC
BOUNDARY CONDITIONS



Calculation of the forces / long-range potentials



Long – range interaction potentials

(e.g. Coulomb):

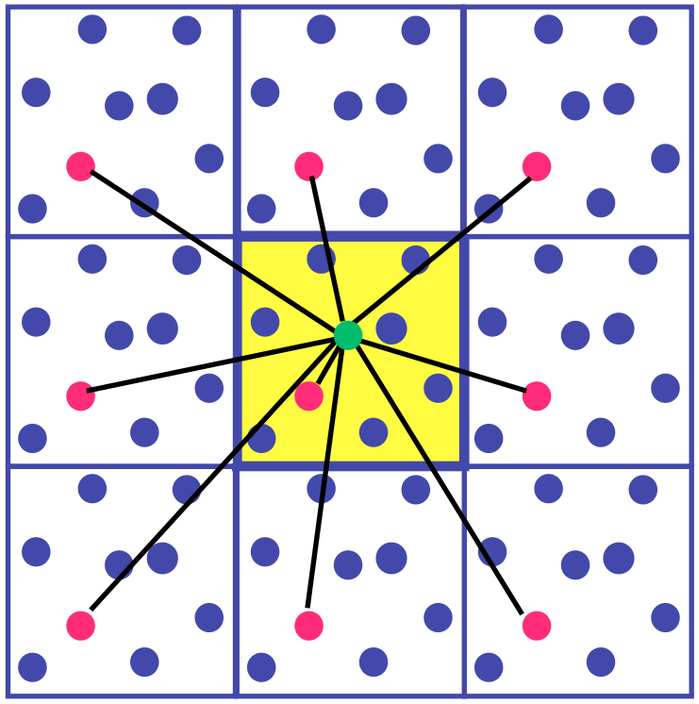
Not possible to find cutoff radius,
"tricks" are needed

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

Possible solutions:

- Ewald summation
- Particle-Particle, Particle-Mesh (PPPM, P3M) method (Hockney & Eastwood)

Calculation of the forces / long-range potentials



The Particle-Particle Particle-Mesh method

Finite size charge clouds

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

$$\int_0^R \rho(r)\mathrm{d}V = Q$$

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

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

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

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)

Calculation of the forces / PPPM method

Particle-Mesh part

1) Assign charges to a mesh: $\rho_m(\mathbf{r})$

2) Calculate 3D FFT: $\hat{\rho}_m(\mathbf{k})$

3) Obtain potential distribution in $\mathbf{k}$-space:

$$\hat{\phi}_m(\mathbf{k}) = G(\mathbf{k})\hat{\rho}_m(\mathbf{k})$$

4) Calculate 3D inverse FFT: $\phi_m(\mathbf{r})$

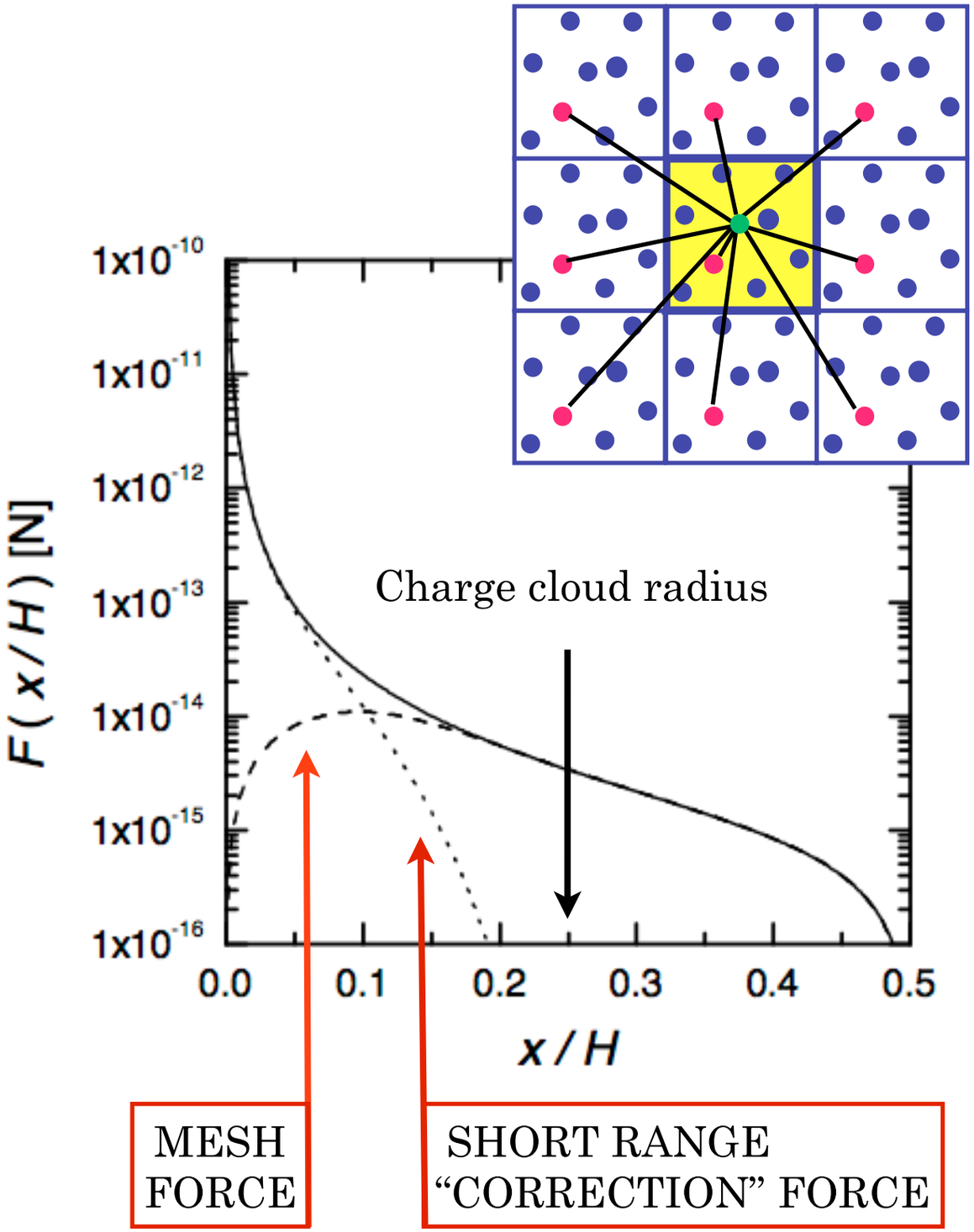
5) Differentiate potential to obtain electric field

6) Calculate force acting on particles via interpolation electric field to particle positions

Particle-Particle part

As done in the case of short-range potentials

Finally add forces before the integration of the equations of motion



Computational methods / MC + MD

Monte Carlo transport calculations

Transport theory

⇩

Interaction of the classical electron gas with the neutral buffer gas
MC

Molecular dynamics simulation

Many-body physics

⇩

Simultaneous many-body interaction within the classical electron gas
MD

$$P_{\mathrm{coll}} = 1 - \exp[-n_g \sigma_t(v) v \Delta t]$$

$$m\frac{\mathrm{d}^2\mathbf{r}_i}{\mathrm{d}t^2} = \sum_{i\neq j}\mathbf{F}_{ij} + e\mathbf{E}$$

Completely first principles
No need for binary approximation of electron-electron "collisions"

$$\sum_{i\neq j}\mathbf{F}_{ij} = ?$$ ✓

- **Zero-D : homogeneous system only**
- **Fix number of particles: ionization treated as conservative process**

Z. Donkó, Physics of Plasmas, 21, 043504 (2014)

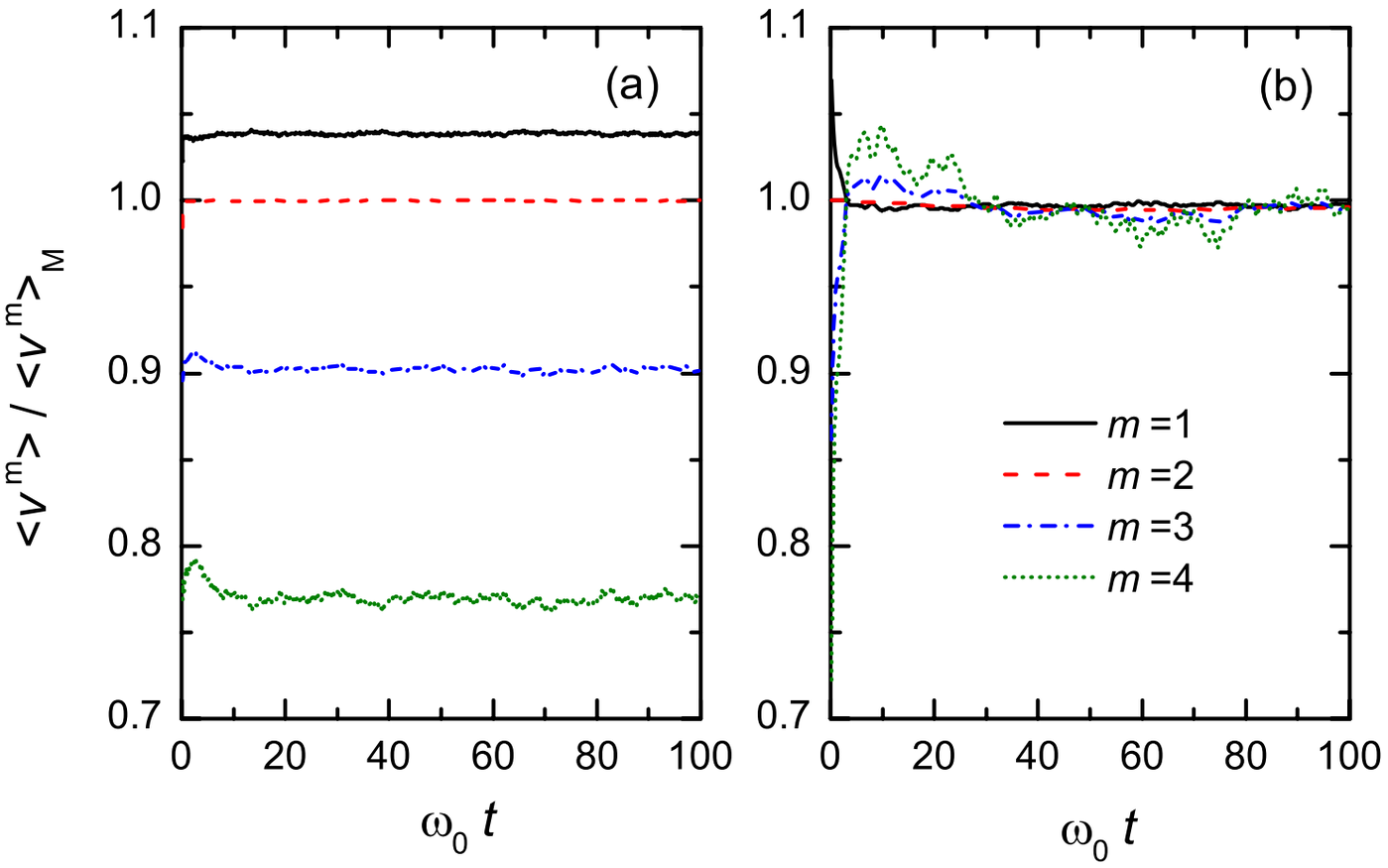
Simulations: monitoring of equilibration

Initial particle configuration:
- random spatial positions
- monoenergetic or Maxwellian energy distribution

Needs to equilibrate !

E/n = 10 Td

η=10^{-6} η=10^{-1} ← Density ratio



Argon

Equilibration is monitored by **moments** of the $f(v,t)$ velocity distribution function

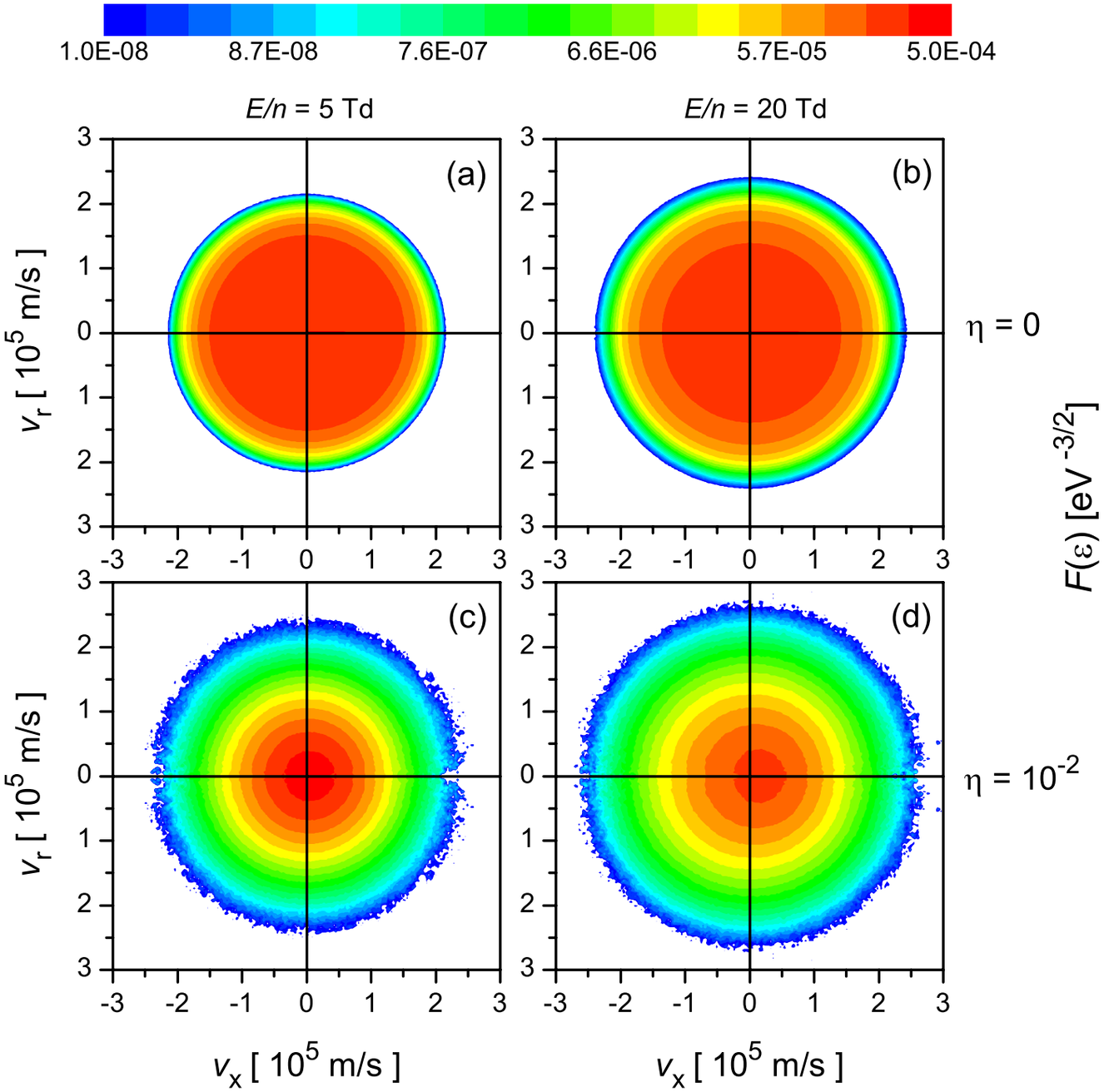
$$\langle v \rangle_M = 2\alpha\sqrt{2/\pi}$$
$$\langle v^2 \rangle_M = 3\alpha^2$$
$$\langle v^3 \rangle_M = 8\alpha^3\sqrt{2/\pi}$$
$$\langle v^4 \rangle_M = 15\alpha^4$$

where $\alpha = \sqrt{2\langle\varepsilon\rangle/3m_e}$

Are we ready to collect data?
How close is the distribution function to Maxwellian?

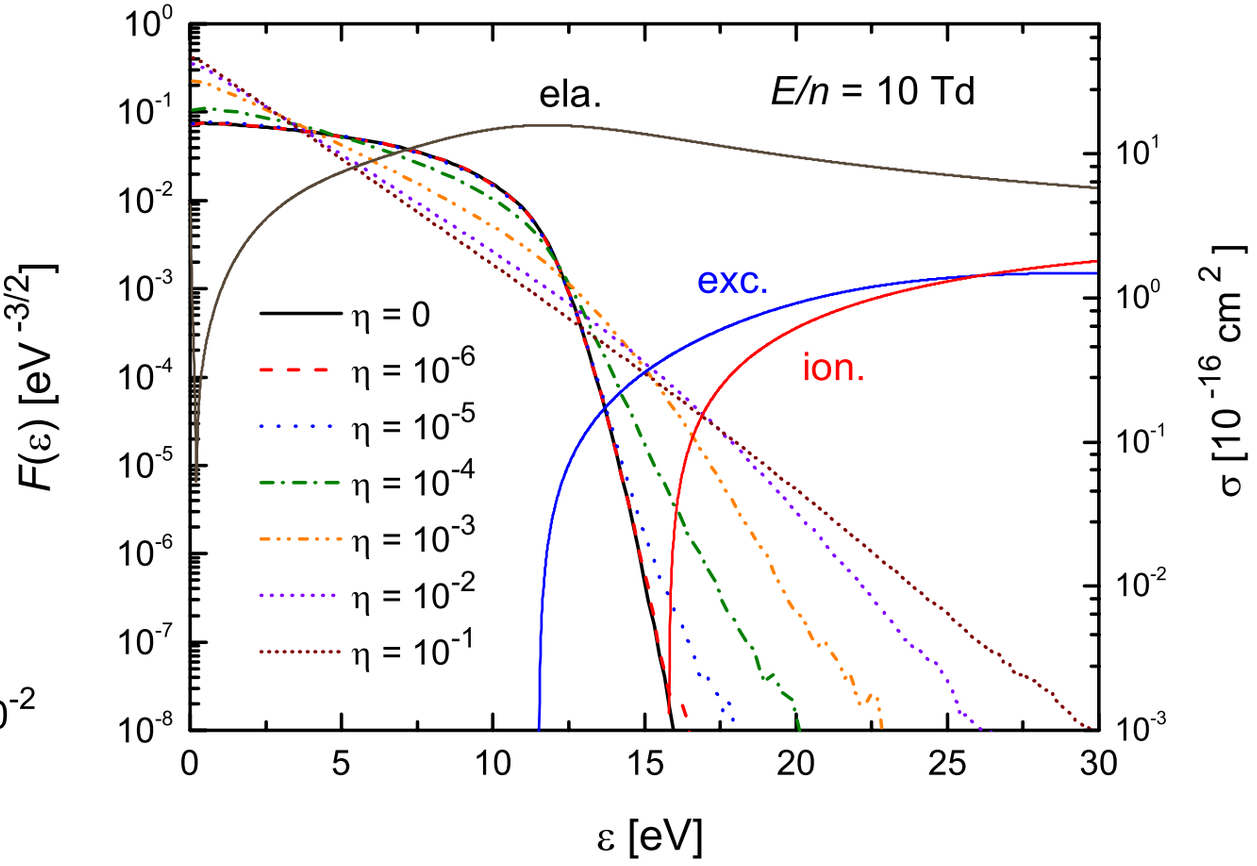
Velocity and energy distribution functions

Velocity distribution functions (VDF)



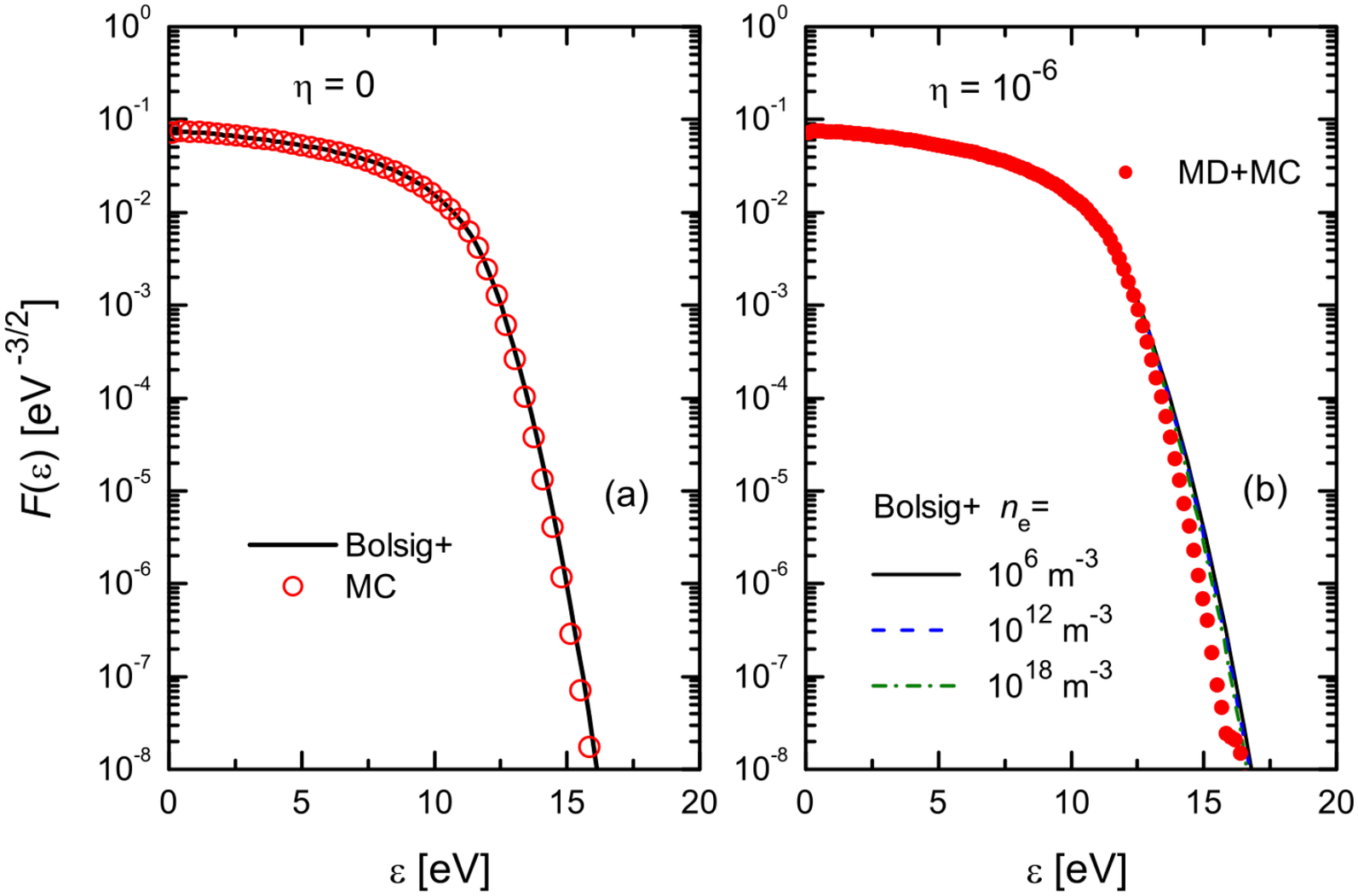
Argon

Energy distribution functions
at different density ratios



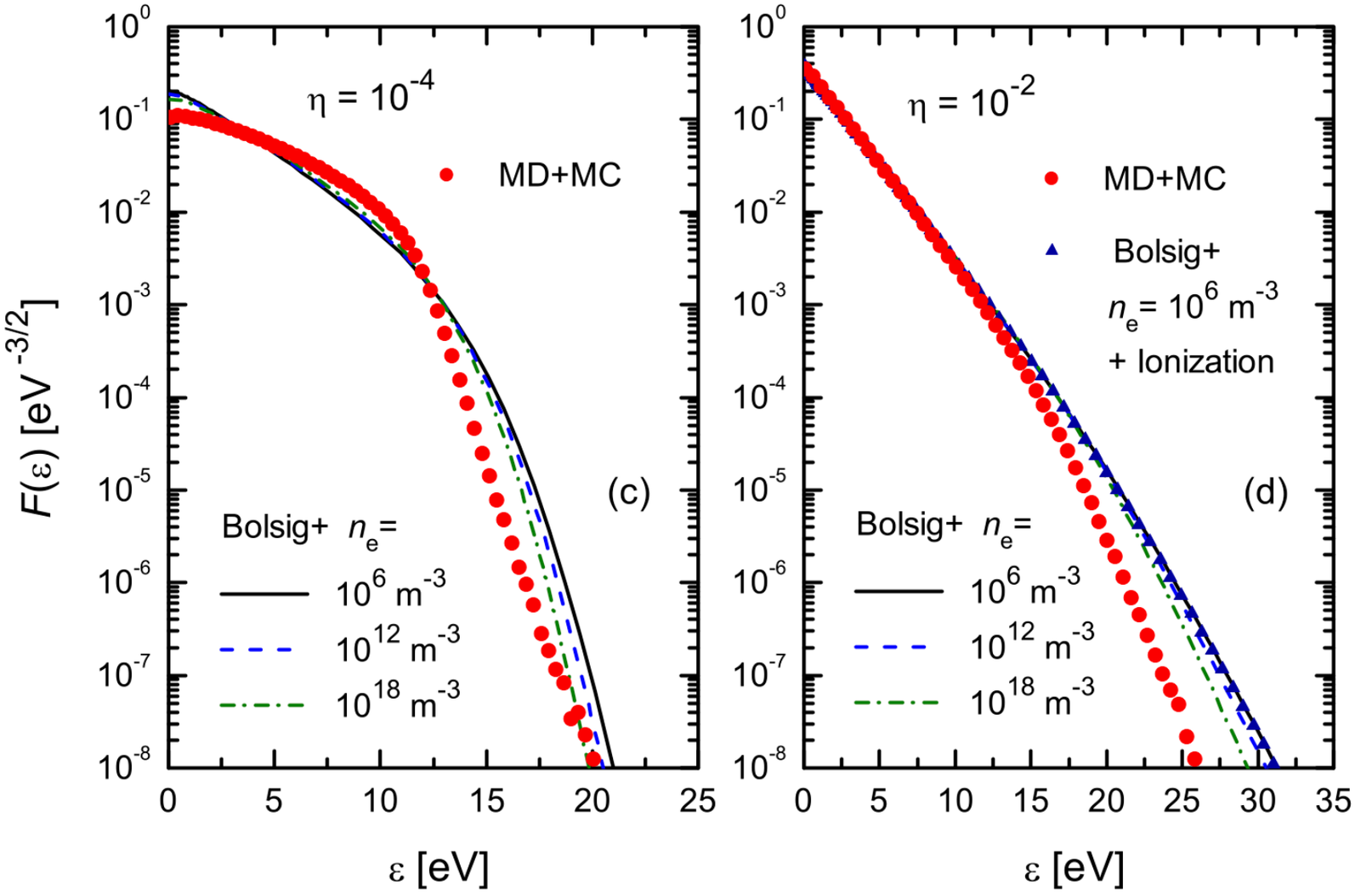
With increasing electron to neutral density ratio the shape changes continuously towards a Maxwellian

Comparison of $F(\varepsilon)$ with 2-term BE solution



- BE solutions: Bolsig+ [G. J. M. Hagelaar and L. C. Pitchford, PSST, 14, 722 (2005)]
- E/n = 10 Td, Argon
- The cross section set of Bolsig+ was modified to treat ionization as a conservative process, just like it is assumed in our particle simulation method
- At η = 0 we use only the MC part. The results are nearly identical with those of Bolsig+

Comparison of $F(\varepsilon)$ with 2-term BE solution

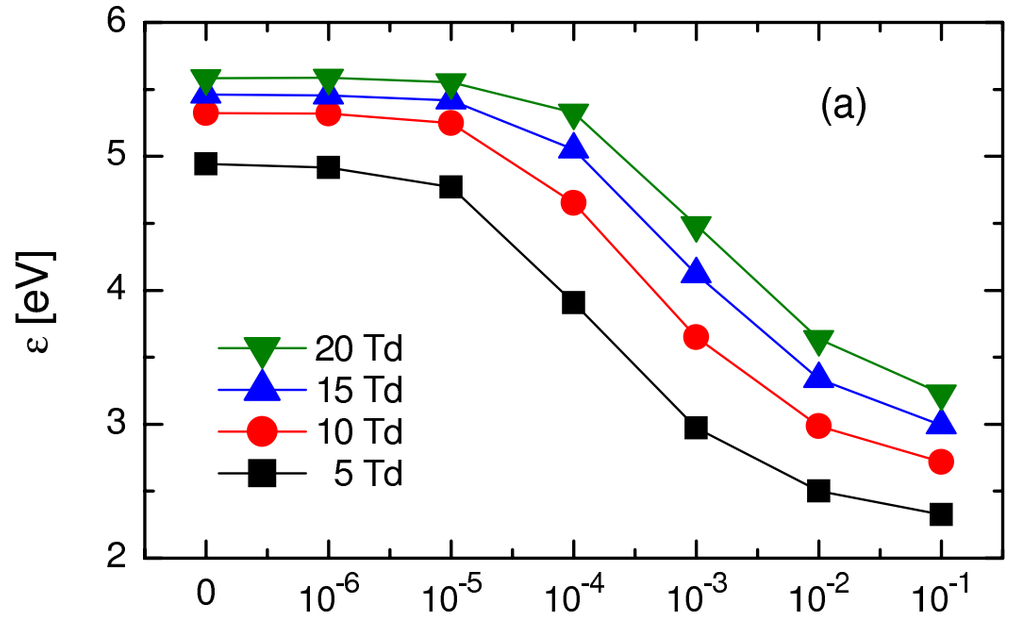


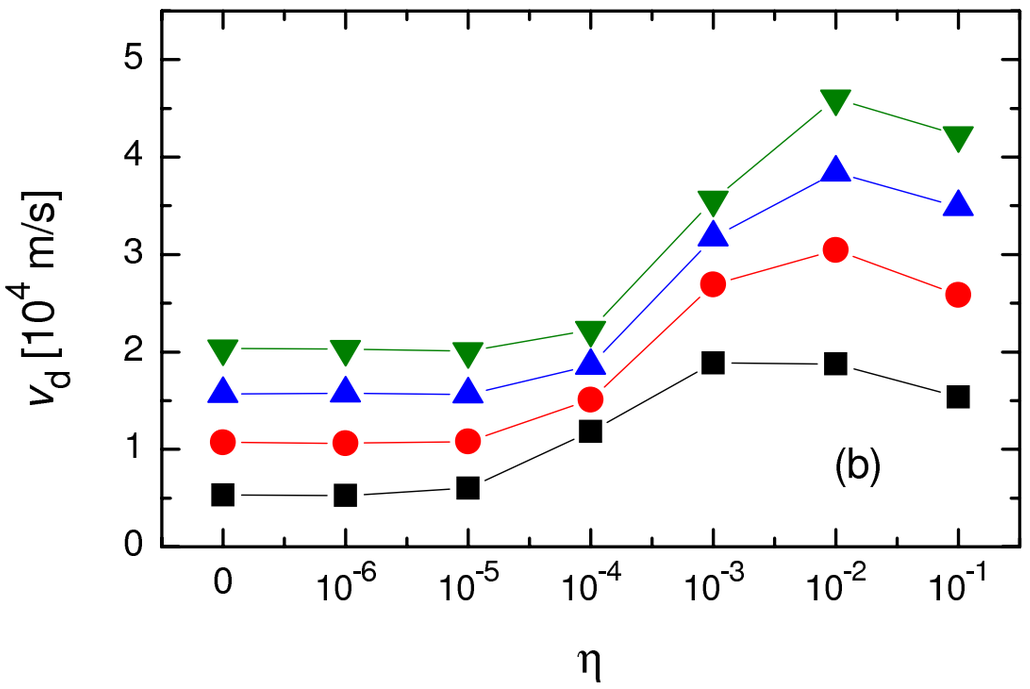
- When Coulomb effects are considered, Boltzmann solvers assume the presence of screening via space charges, and thus, use the absolute value of the electron density as an input parameter. The results exhibit a weak dependence on the electron density
- Significant deviations indicate possible issues with the presently available binary collision treatment of Coulomb collisions in MC and BE solutions
- The effect of treating the ionization as a non-conservative process was tested with Bolsig+

Transport coefficients

The changes of the VDF influence the transport parameters:

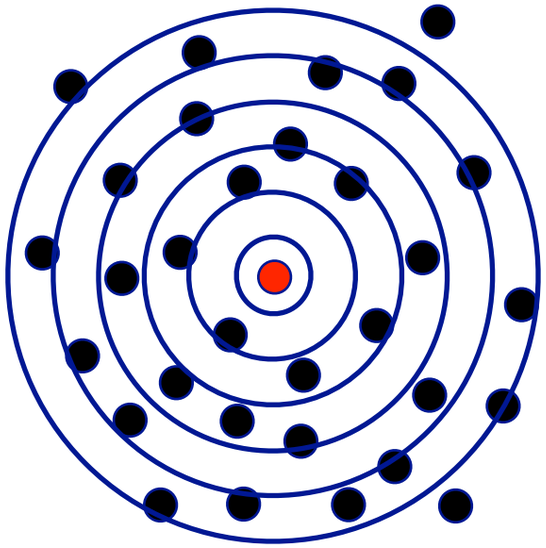
- The mean energy of the electrons decreases with increasing density ratio, significant changes take place above $\eta \sim 10^{-5}$
- The drift velocity, on the other hand, increases with increasing η, but beyond a certain, high value of η (that depends on E/n), v_d starts to decrease
- The changes of the VDF influence the reaction rates as well. Increasing population of high energy electrons results in less elastic and more inelastic collisions - *limitation* of the model (ionization)!

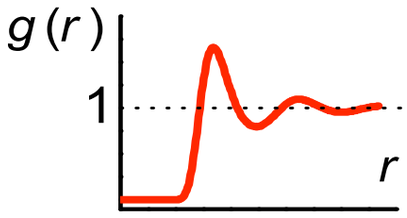




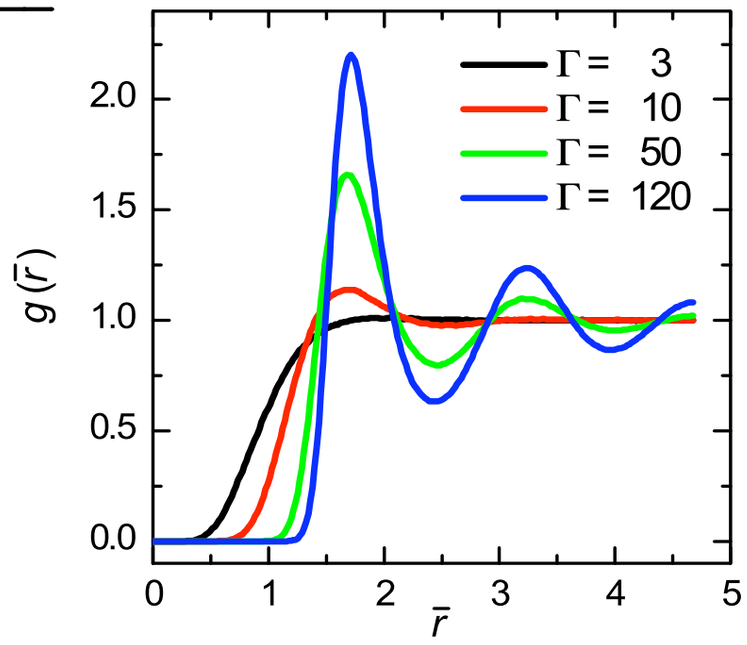
Pair correlations between electrons

Pair correlation function

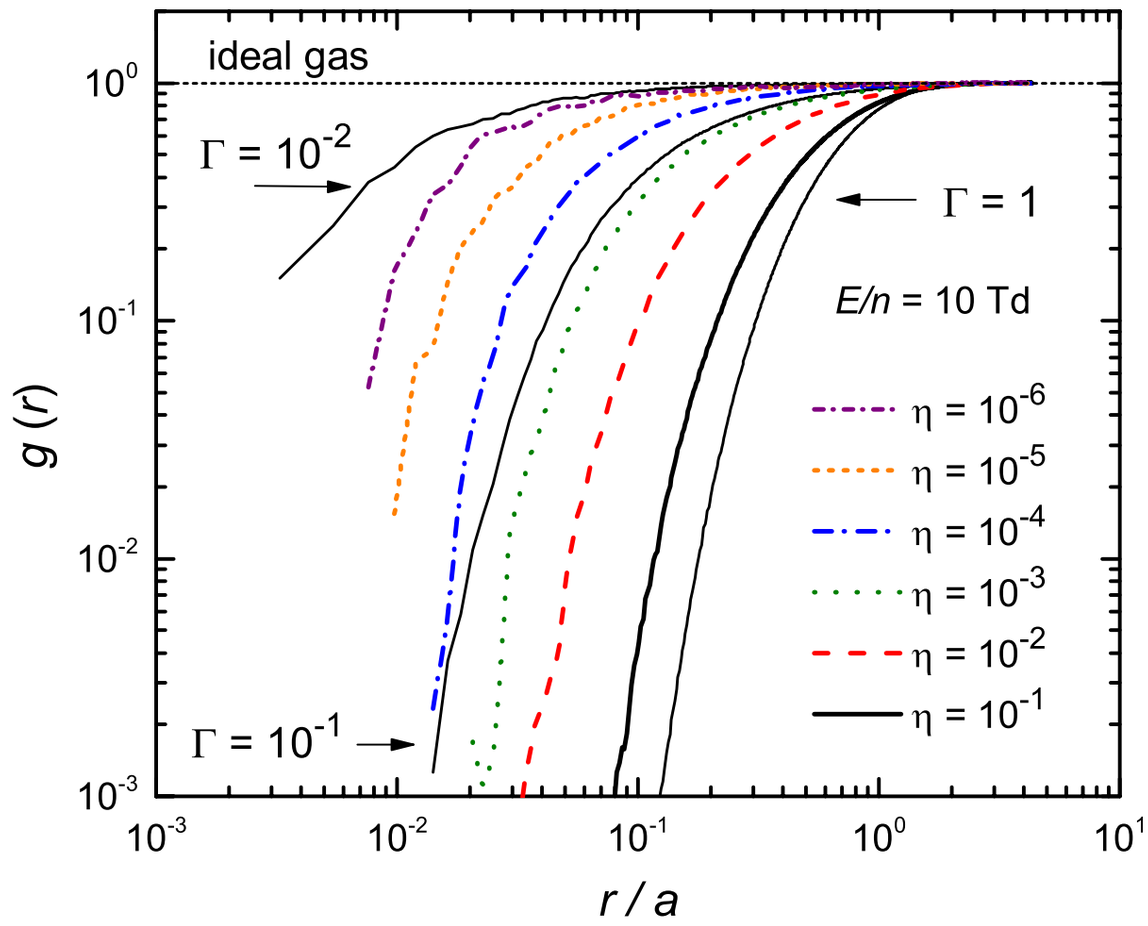




Relative density of particles around a test particle, with respect to the ideal gas



Pair correlation within the electron swarm



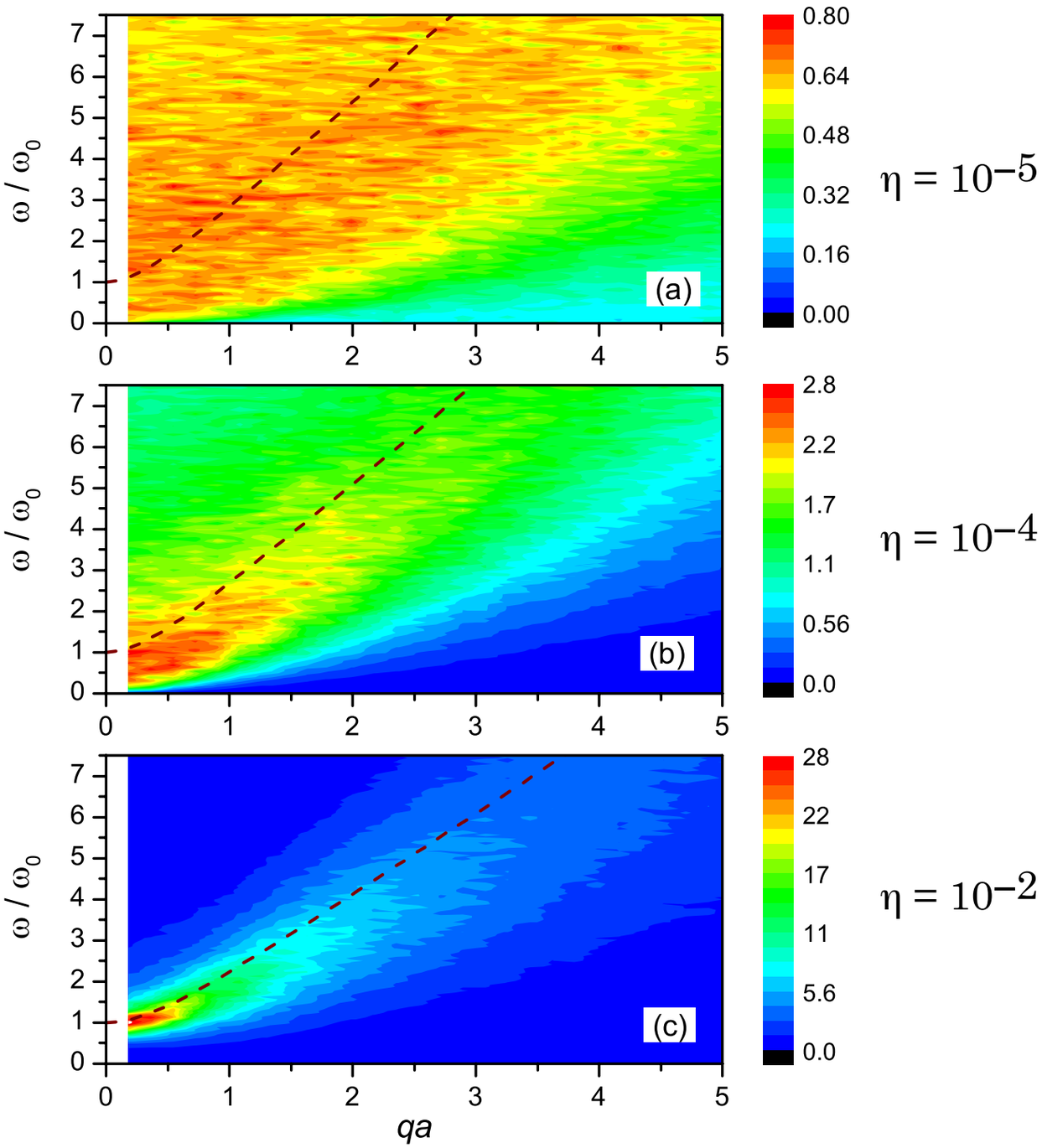
Reference system: *one-component plasma*

$\Gamma = e^2/(4\pi\epsilon_0 a k_B T)$ coupling parameter

$a = (3/4\pi n_e)^{1/3}$ Wigner-Seitz radius

Electrostatic waves within the swarm

Current-current fluctuation spectra $L(q, \omega)$



Microscopic particle current (*Fourier transform in space*):

$$\lambda(q,t) = \sum_j v_{jx}(t) \mathrm{e}^{iqx_j(t)}$$

Current-current fluctuation spectrum (*Fourier transform in time*):

$$L(q,\omega) = \frac{1}{2\pi N} \lim_{\tau\to\infty} \frac{1}{\tau} |\lambda(q,\omega)|^2$$

- at low η the energy is spread widely in the (q, ω) plane
- with increasing electron to neutral density ratio a pronounced **collective mode** develops gradually
- at $q \to 0$ the mode frequency equals the plasma frequency, $\omega = \omega_0$
- with increasing wave number ω increases, following closely the **Bohm-Gross dispersion relation of warm electrostatic waves** (indicated by the dashed lines):

$$\omega^2 = \omega_0^2 + \frac{3k_B T_e}{m_e} q^2 \qquad \omega_0^2 = n_e e^2/\varepsilon_0 m_e$$

Summary and outlook

- **An approximation-free method for the description of the motion of electrons in a background gas, under the influence of an external electric field and electron-electron interactions, at arbitrary ratio of electron to neutral density** [Z. Donkó, Physics of Plasmas 21, 043504 (2014)]
- The method is based on a **combination of the classical Monte Carlo (MC) technique and a Molecular Dynamics (MD) method**:
  - the MD simulation describes the (multi-particle) interactions within the electron gas
  - the interaction of the electron gas with the background gas is handled by the MC procedure
  - VDF, energy distribution function, transport parameters, rate coefficients + pair correlations and collective excitations (waves)
- **Future directions**:
  - warm gas ✓
  - (non-conservative) ionization
  - time-dependent problems
  - time-varying fields
  - ....

Thank you for your attention

This talk can be downloaded from
http://plasma.szfki.kfki.hu/~zoli/presentations.cgi