Simulations of Ion Stopping Power in Warm Dense Matter

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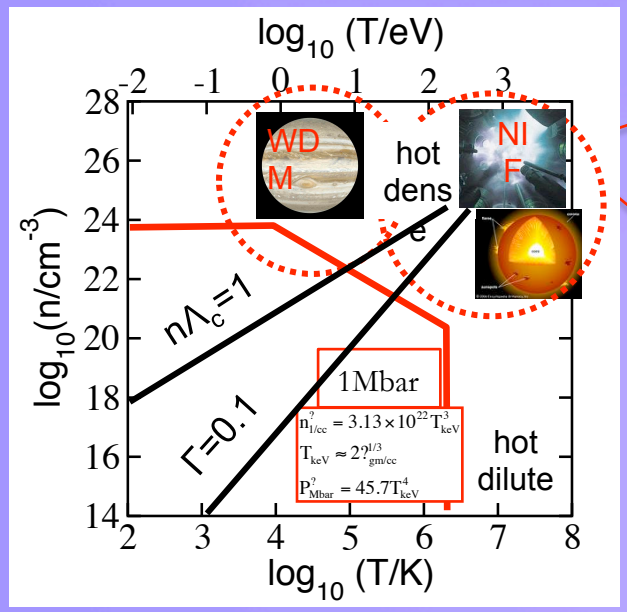
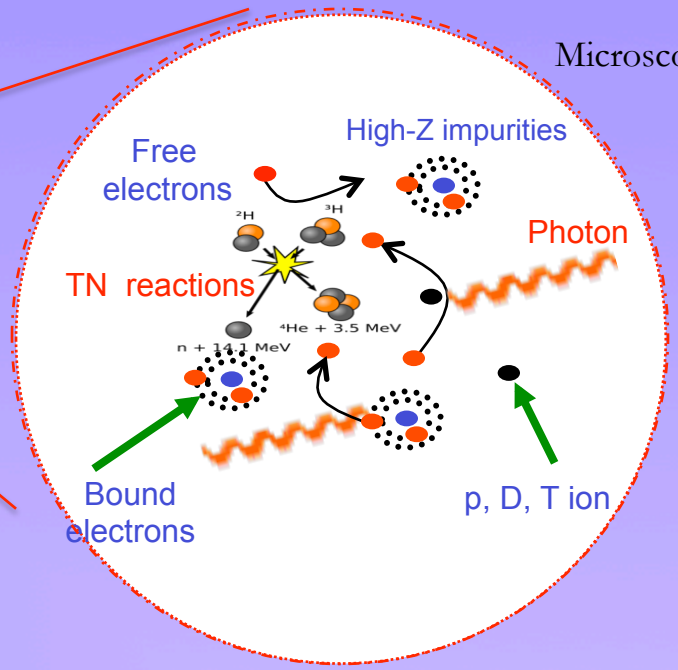
Our starting point is augmenting molecular dynamics to model hot dense, burning radiative plasma: Overview

Macroscopic



Microscopic



✓MD is ideally suited for fully-ionized, nondegenerate plasma: Coulomb interaction is included to all orders

Long range = incoherent, classical $\lambda_{deBroglie} < r_s$
Short range = coherent, quantum $V_{coul}(r\text{->}0) > kT$

! Microscopic processes are a mix of classical and quantum physics

We want to develop and compare mixed quantum/classical simulations to analytic models and experiment*

*** = e.g. stopping power in WDM**

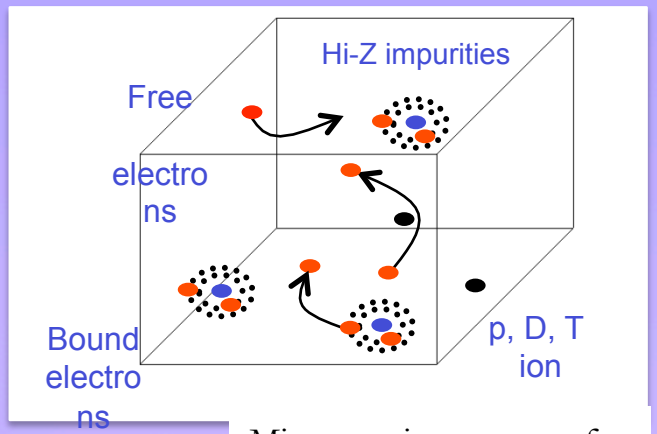
We must go beyond classical MD while assessing the reliability of our modifications: Specifics

1. High-temperature limit allows classical trajectories (Coulomb molecular dynamics) *

2. High-Z impurities introduce strong coupling physics, QM bound states *
   1. Quantum phenomena will be introduced via coextensive Monte Carlo models
      1. Bound core states
      2. dense plasma effects, e.g. continuum lowering
      3. Dynamical core excitation, ionization, recombination

3. Compare to theory, analytic models
   a) Like-charged projectiles in OCP*
   b) Projectiles in H, no bound states (energy split)
   c) Targets with high-Z impurities, and WDM* (atomic physics)

4. Compare to experiments
   a) Jupiter Laser Facility proton stopping in warm dense C *



Microscopic processes for the stopping problem

*** = stopping work to date**

We Are Interested In Two Issues for Stopping

1. What are the current limitations for kinetic theories of stopping power? (HDRP)
   a) Strong, correlated collision problem:
      strong beam-target interaction with collective excitations is an unsolved problem
   b) Here, simulations provide non-perturbative (numerically converged) results,
      using idealized models that have no uncertainties (e.g., repulsive Coulombic OCP)

2. What is the stopping behavior in real matter?
   a) strong electron-electron interactions (dynamical local field corrections) (WDM)
   b) degeneracy, Pauli blocking in electronic subsystem (WDM)
   c) strong ion-ion interactions (strong coupling liquid) (WDM, HDRP)
   d) strong electron-ion interactions (bound states, excitations, ionization by projectile) (WDM, HDRP)

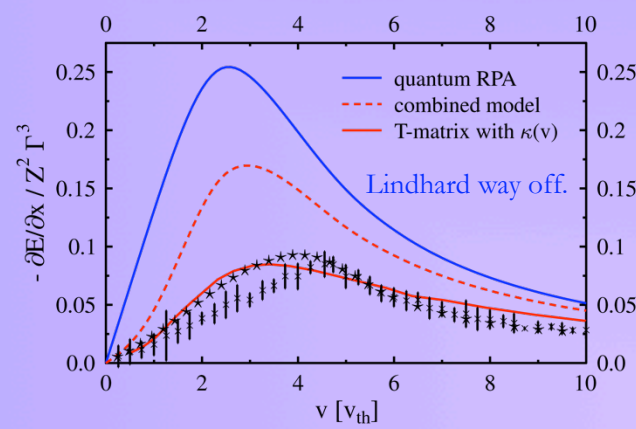
Issue #1: Tests of Kinetic Theories

- Many stopping power models (e.g., Lindhard) are based on perturbation theory
- Classical model is most difficult (quantum stopping naturally convergent)
- Here, we perform simulations using pure classical dynamics
  - in regimes where perturbation theory fails
  - disallowing formation of any bound states in target or with projectile

Our simulation model:

1. classical electron one component plasma: no discrete ions in target = no atomic states
2. projectile charge is negative, so that no bound states form on the projectile
3. projectile charge is large, to enhance beam-target coupling
4. use molecular dynamics to resolve all trajectory information in strongly coupled system (interactions to all orders)

A great deal has been learned from previous studies, but state of the art MD has improved meanwhile

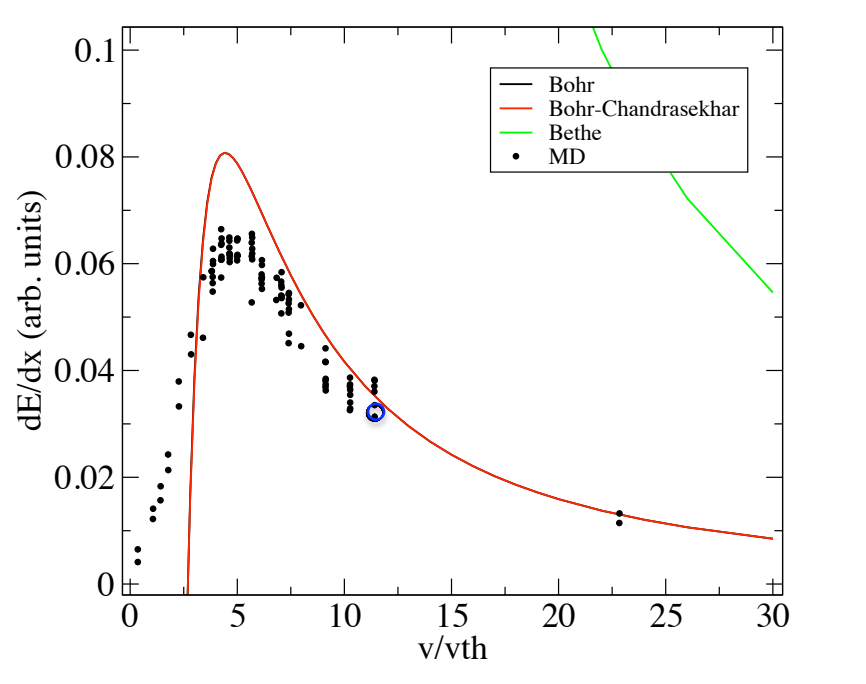


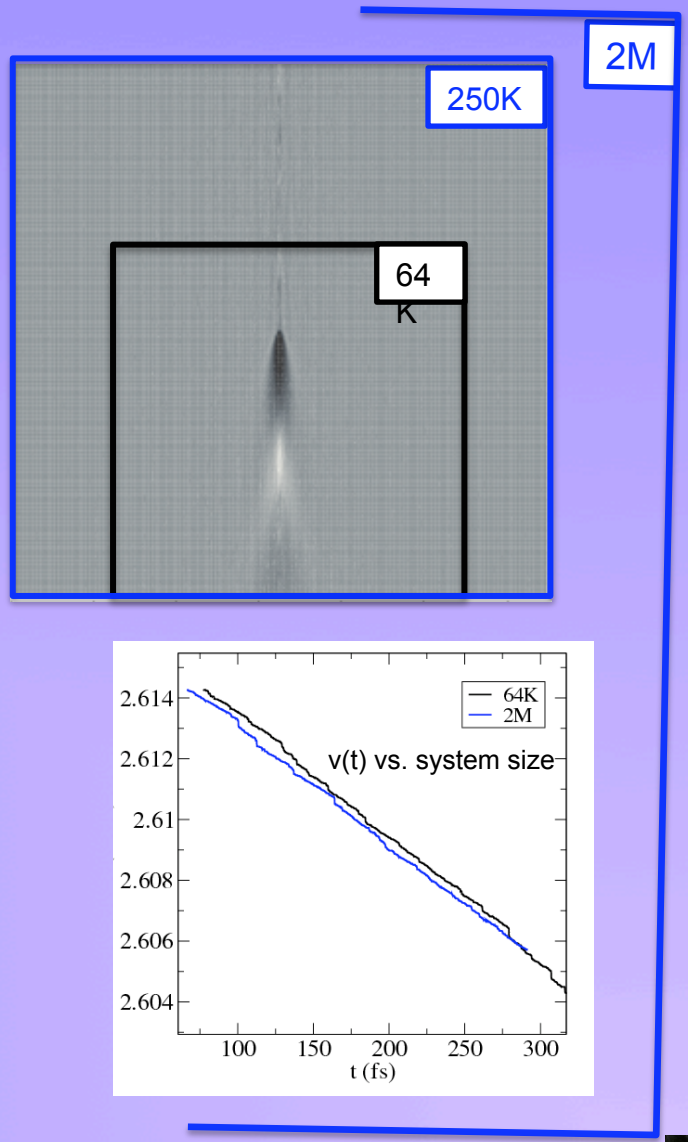
MD for OCP: Coulomb interactions can be evaluated effectively free from finite system-size errors.

Most early MD used 10^3-10^5 particles
We use $6X10^4$ and $2X10^6$, up to $2X10^9$ charged particles

Our finite size effects are under control
(e.g., dE/dx for 64K particles • is similar to 2M ○)

dE/dx vs. v for Z=-10 in e^- gas, Γ=1





We apply a weak thermostat to the target to control heating.
A fast projectile travels 1000-2000 Å in relaxation time τ

OCP vs. Theoretical Models, dielectric formulation

There are analytic expressions for stopping and straggling in the linear response regime

$$\left\langle \frac{dE_b}{dt} \right\rangle = -\frac{2(Ze)^2}{\pi v^2} \mathrm{Im} \int_0^\infty \frac{dk}{k} \int_{\frac{\hbar^2 k^2}{2m_b} - kv}^{\frac{\hbar^2 k^2}{2m_b} + kv} d\omega \left(\omega - \frac{\hbar k^2}{2m_b} \right) \mathrm{Im}\, \varepsilon^{-1}(k,\omega) n_B(\omega)$$

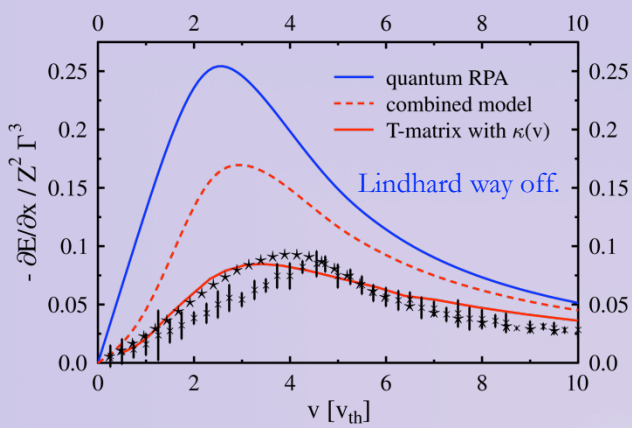
RPA and local-field corrected version have been studied

High temperature (classical limits), relevant to Coulombic MD

$$\frac{dE}{dx} = -\frac{(Ze)^2}{\pi v^2} \mathrm{Im} \int_0^{k_{\max}} \frac{dk}{k} \int_{-kv}^{+kv} d\omega \frac{\omega}{\varepsilon(k,\omega)}$$

(Integrals of V_{coul} are not convergent here)

Weak-coupling, linear response is inadequate for large Z



Gericke, private comm.

OCP vs. Theoretical Models: The combined method is well-known

Strong coupling scattering can be evaluated by the T-matrix for static screening

$$\frac{dE_b}{dx} = -\frac{1}{(2\pi)^2\hbar^3}\sum_{species\,c}\frac{m_c^{\ 2}}{m_{bc}^3}\frac{n_c\Lambda_c^3}{v}k_BT\int_0^\infty dp\,p^3Q_{bc}^T(p)$$

$$\times\left(p_-Exp\left(-\frac{m_cv_-^2}{2K_BT}\right)-p_+Exp\left(-\frac{m_cv_+^2}{2K_BT}\right)\right)$$

In terms of thermal deBroglie wave length Λ, transport cross section Q, …

Finally, static higher-orders, and dynamic linear stopping can be combined, Gericke and Schlanges, Morawetz and Ropke, (Gould and deWitt)

$$\frac{d}{dx}\langle E_b\rangle = \frac{d}{dx}\langle E_b\rangle^{T-matrix} + \frac{d}{dx}\langle E_b\rangle^{RPA} - \frac{d}{dx}\langle E_b\rangle^{staticBorn}$$

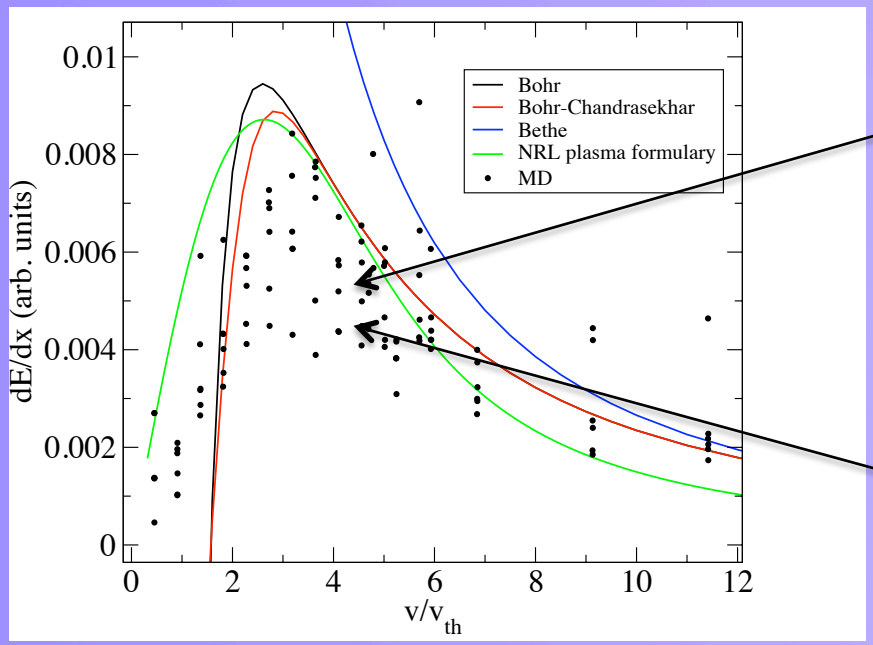
MD simulations provide fluctuation statistics

MD includes detailed statistics on scattering, transport and correlation functions

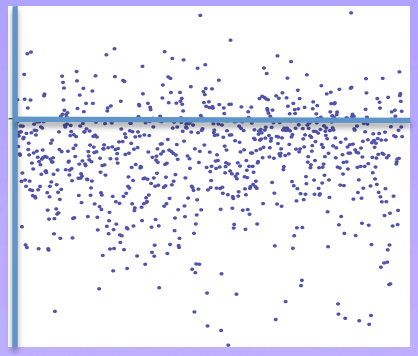
e.g., on straggling and blooming

We are in the process of comparing to analytic models

dE/dx vs. v for Z=-2 in e^- gas, Γ=1

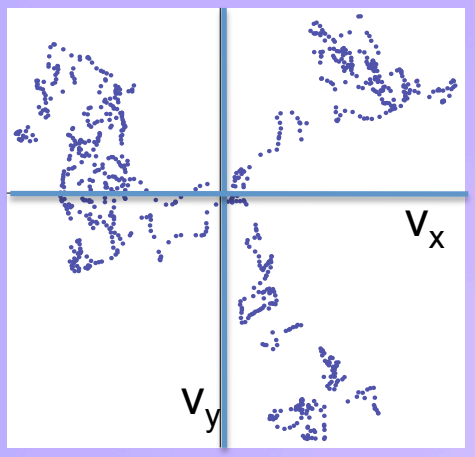


δv_{Long} vs. t



0

Hard collisions contribute to straggling

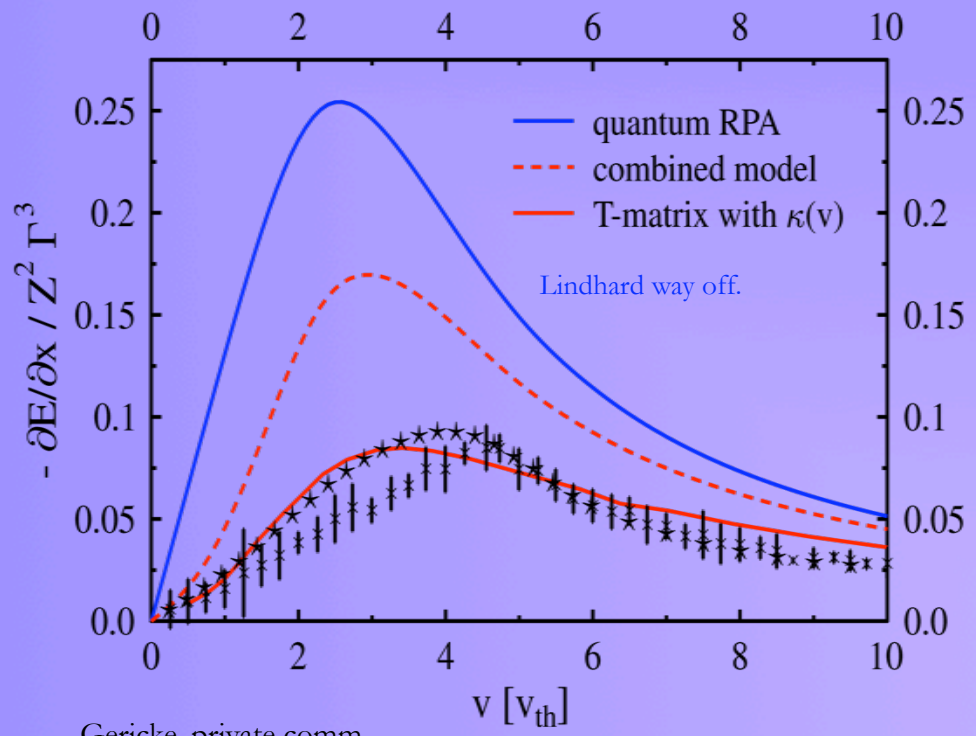


$\int \delta v_{trans}$

Avg~0

Finite size effects may be more severe for fluctuations from the average

OCP vs. Theoretical Models



Gericke, private comm.

Stopping includes linear drag regime for slow projectiles

Peak energy transfer for $v \sim v_{th}$

Asymptotic fast projectile stopping $\sim \ln v/v^2$

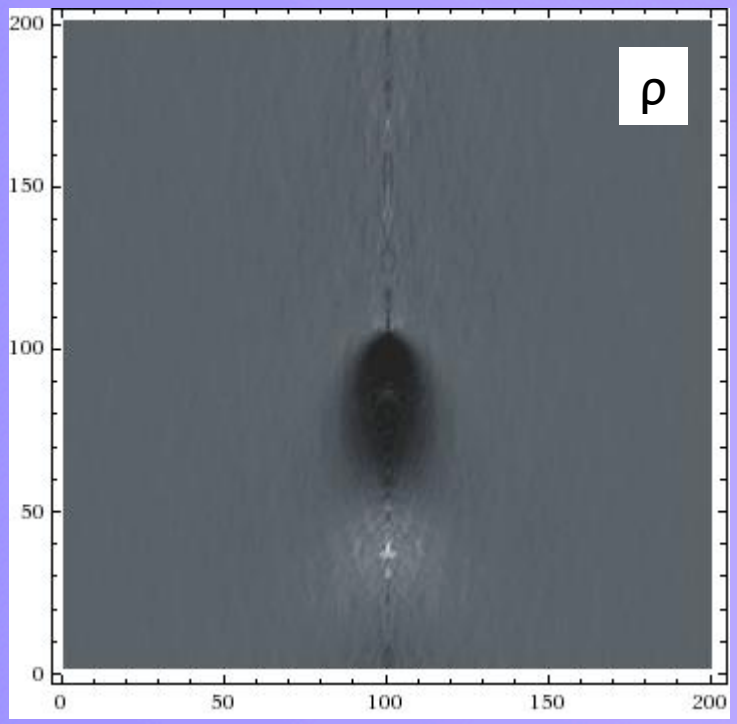
We are working on local field corrected dielectric response calculations (Ichimaru, "fluctuation approach")

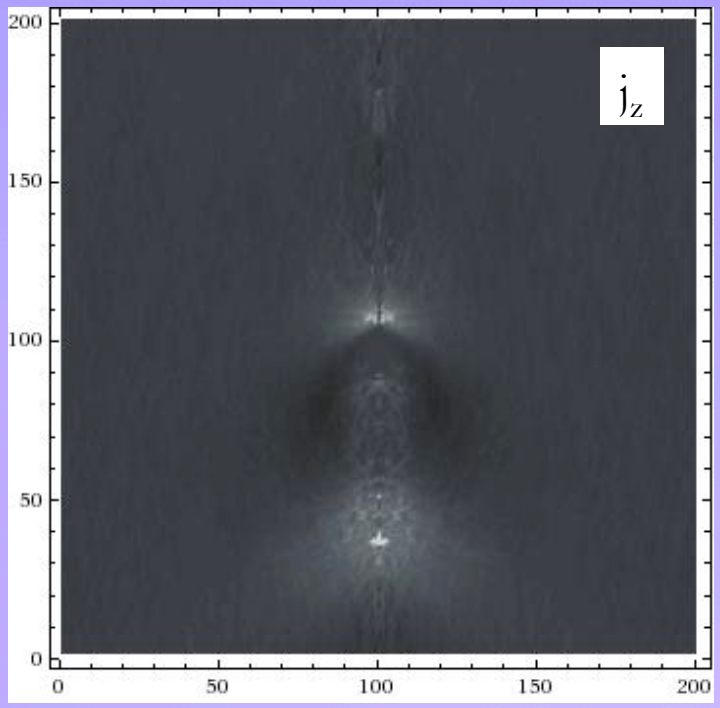
T-matrix with velocity-dependent screening also gives good agreement for strong coupling

MD simulations include details of the velocity-dependent dynamical screening in strong coupling

Pure repulsive Coulomb, Z=-10, Γ=1 OCP $v/v_{th}=4.3$

Time averaged over 10^2 fs, in strong-coupling limit





Current density reveals both collective turbulence and single particle excitations

Issue #2: Atomic physics and degeneracy effects in Warm Dense Matter

- Stopping power in warm dense matter is a difficult problem
  - Strong coupling among all species, quantum mechanics manifests in:
    1. Screening
    2. Atomic states in target
    3. Pauli blocking in bound-free, free-free excitations by projectile
    4. Transient bound states of projectile

Thus, we are in a serious bind: we need to solve the dynamical quantum many-body problem!

Our strategy:

- Assume *long-range* dynamics are dominated by classical Coulomb scattering
  (e.g., the plasmon energy is correctly predicted by classical physics)
- Assume that the remainder is described by *short-range* quantum corrections
  use effective potentials for particle trajectories and Monte Carlo for quantum transitions

This creates a viable "dynamical QMD" that, while being imperfect, is tractable, testable, and improvable.

What is our stopping model for real ions?

Theory goal is a method with quantum bound states and transitions,
plus classical nondegenerate free electrons

For a first look, we just want to examine computational feasibility

We have applied classical MD to stopping for a C^{6+} model with classical bound electrons

We replace singular 1/r binding with an effective quantum-corrected electron-ion potential
use existing high temperature potentials, sufficient to prevent singular binding

MD timestep is set by fastest particle, usually the projectile

This problem is not appreciably more difficult to simulate than OCP

We are modifying the effective potential and core dynamics subsequently

What does classical dynamics predict for real ions?

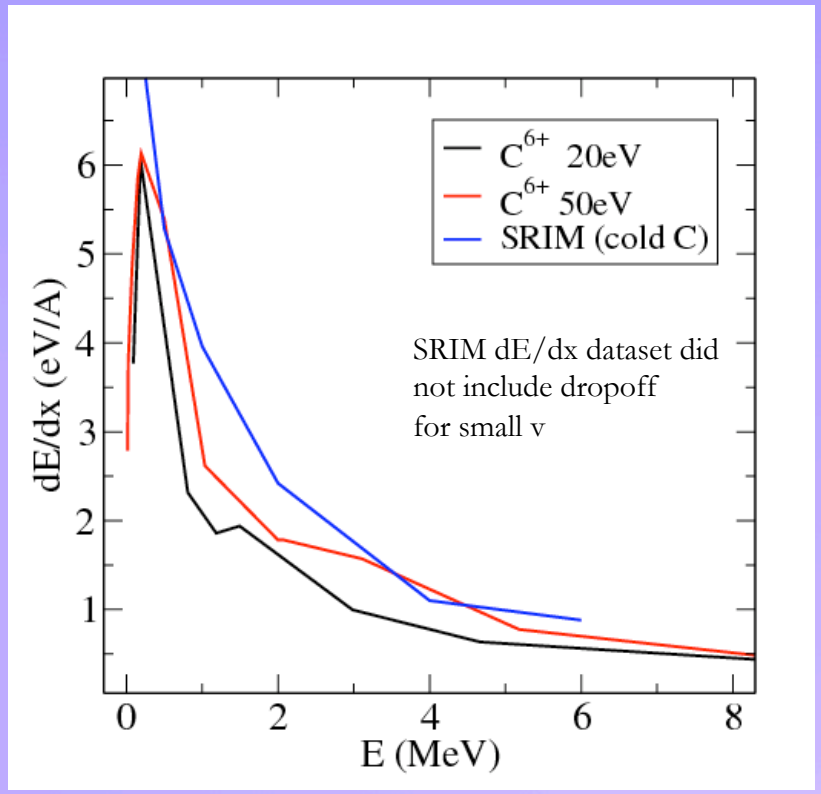
We tried several models for C ions, treated as point effective charges

This includes modeling C^{3+} with a rigid core as a point charge and modeling fully stripped C with classically-bound electrons

E.g., simulate 4000 C^{6+} atoms, 24000 electrons
Graphite density, 2.26 g/cm^3
Single incident proton projectile

At 50+ eV free electrons are not degenerate
Corrections will mainly be to core dynamics and scattering properties

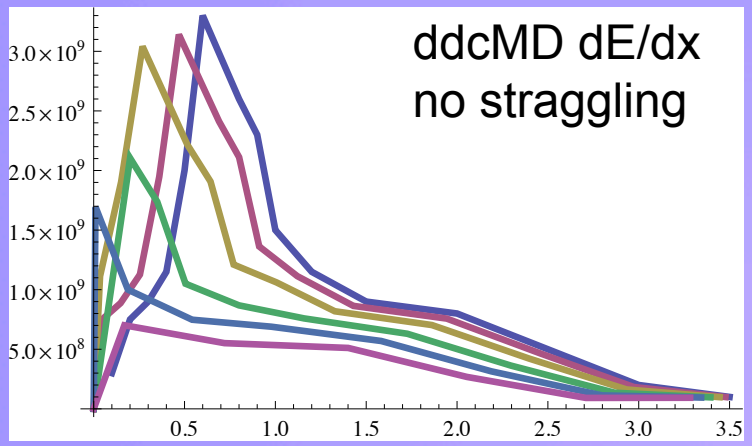
At 20 eV, degeneracy effects are important
These are not included in classical free electron trajectories

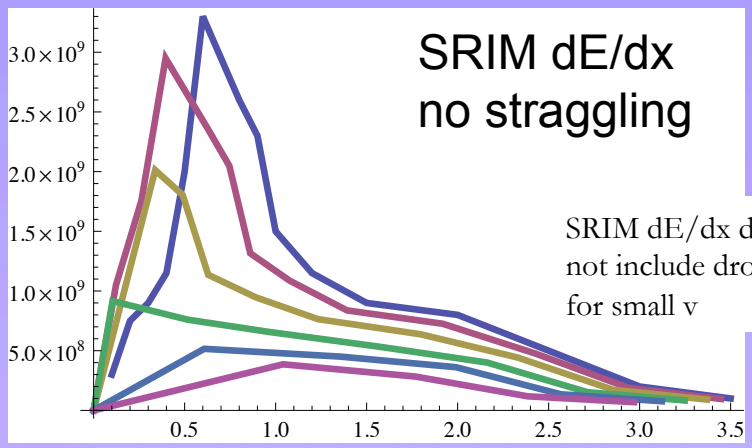


Simulations are qualitatively consistent with expectations
Suggest that quantum atomic corrections will be manageable

Proton in stopping in warm C is underestimated compared to SRIM, for cold C.

Estimated spectrum from 'Shot 54':
Simulated transmission through 2.5, 6.0, 11.5, 17, 24 μm C foil

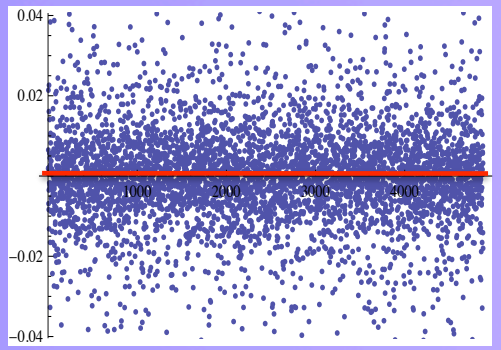




SRIM dE/dx dataset did not include dropoff for small v

Blooming is prominent

Transverse δv
δt = 5000 timesteps



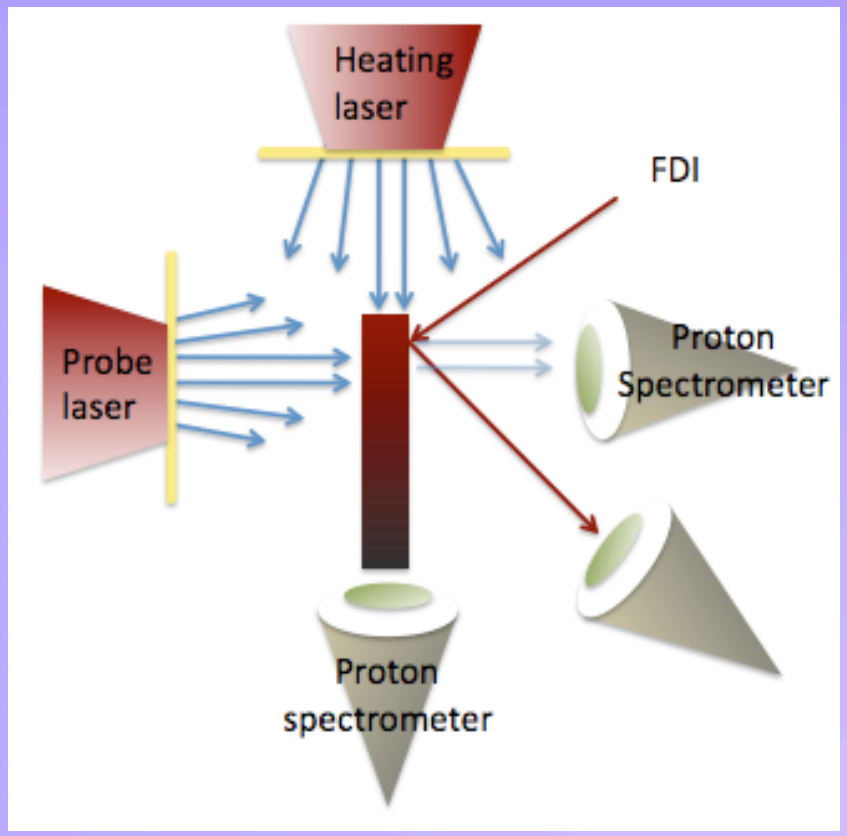
500keV proton loses 10% energy in 1μm.
Variance in transverse v equals a 1-2° deflection.

2MeV proton loses 1% energy in 1μm, sees a 0.2° deflection.

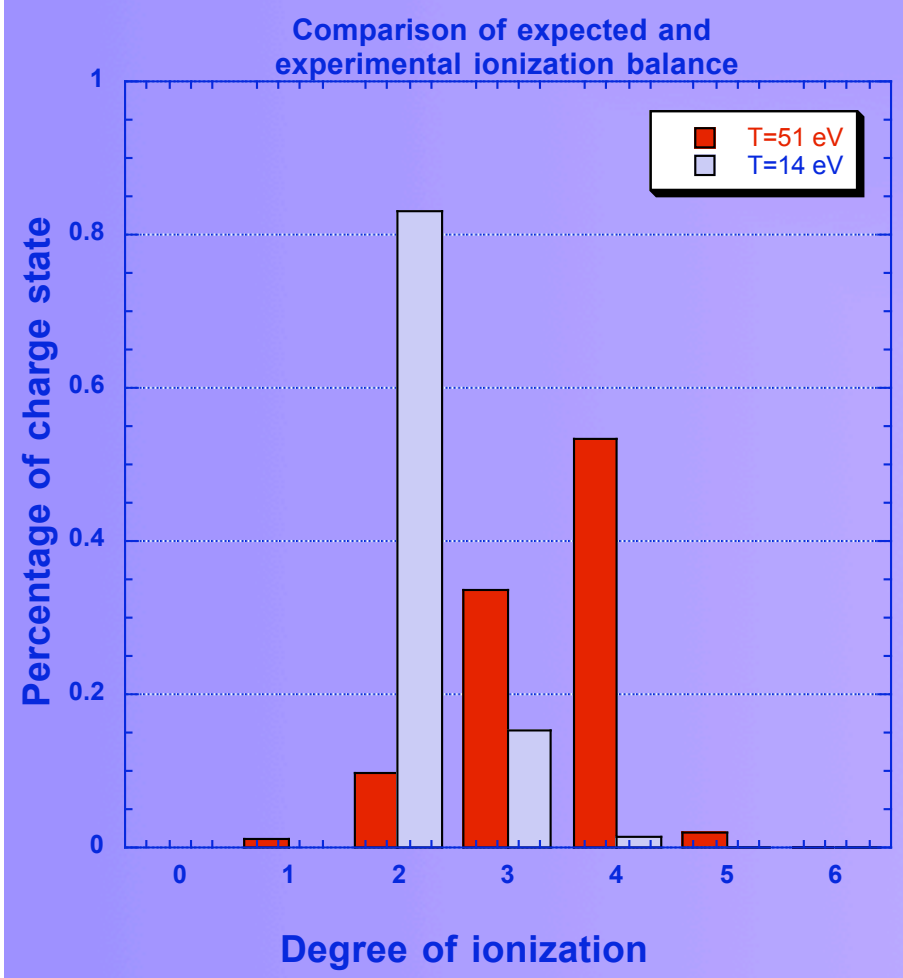
We are performing experiments on C in the WDM regime

Experimental goal is warm dense regime, nondegenerate free electrons, simple core states, e.g. He-like carbon

- Protons heat edge-on
  - Typical heating energy~130 J
  - Typical probe energy~20 J
- Proton spectrometer measures heating spectrum
  - Spectrum is used to infer temperature
- FDI measures expansion of critical surface
  - Expansion velocity is used to infer temperature



The ionization dynamics of the carbon is critical to understanding the stopping power



$$S = \sum_n B\left[\left(1-\bar{Z}/Z_a\right)\ln\Lambda_{bn}\right] + \left(\bar{Z}/Z_a\right)\ln\Lambda_f$$

$$B = 4\pi e^4 N Z_a / mV^2,\ \Lambda_{bn} = 2mV^2/I_{zn},\ \Lambda_f = 2mV^2/I_f,\ Z_a = \text{atomic number}$$

$$N = \text{plasma density},\ V = \text{proton velocity},\ \bar{Z} = \text{average ionization}$$

$$I_{zn} = \text{ionization potential},\ I_f = \bar{Z}e^2/\lambda_D$$

- Ionization balance calculated using FLYCHK
  - Solid density
  - Stewart-Pyatt continuum lowering

•The bound electron stopping is •dominated by the C^{2+} charge state.

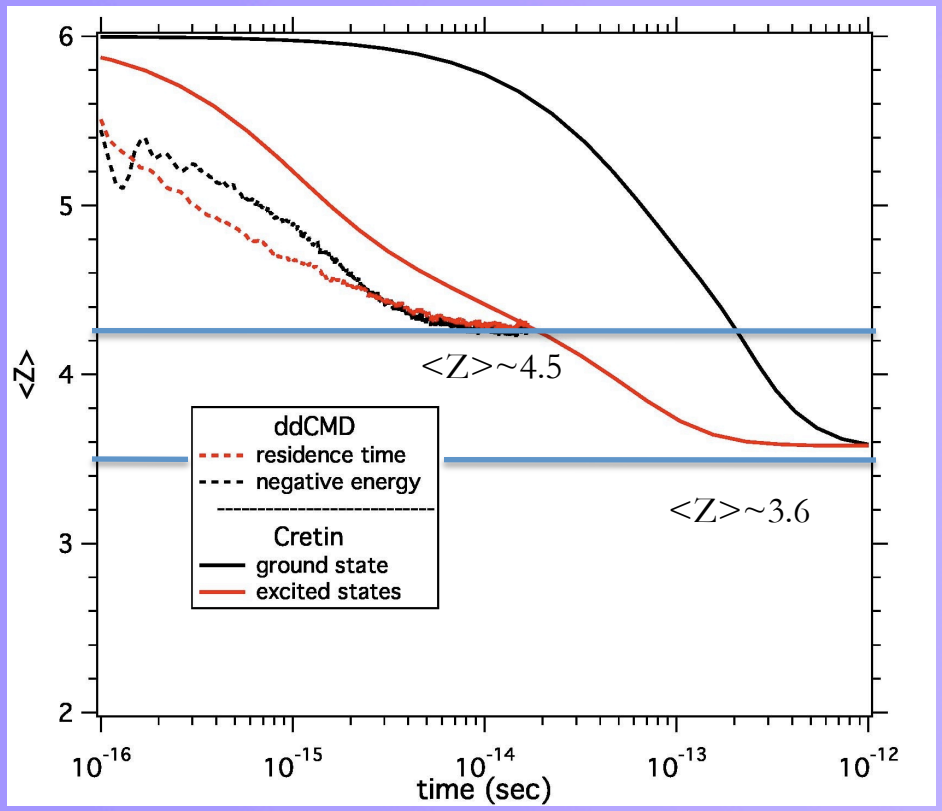
- For our plasma we have:

$$\Gamma_{ii} \approx 5,\ \Gamma_{ie} \approx 2,\ \vartheta_{Deg} = \frac{\theta_{Fermi}}{\theta_{Thermal}} \approx 1.19$$

Free electron stopping is in a partially degenerate gas

Warm Dense Carbon Results: Classical MD

For the DunnBroyles-Deutsch potential:
Start with ionized C 50eV run to equilibrium, what is Z(t)
Use 2 definitions for bound state: instantaneous negative energy or residence time (orbit time)
Classical dynamics predicts too fast recombination rate, equilibrium <Z> ~ 4.5
Cretin dense plasma simulation predicts slower recombination to <Z> ~3.5



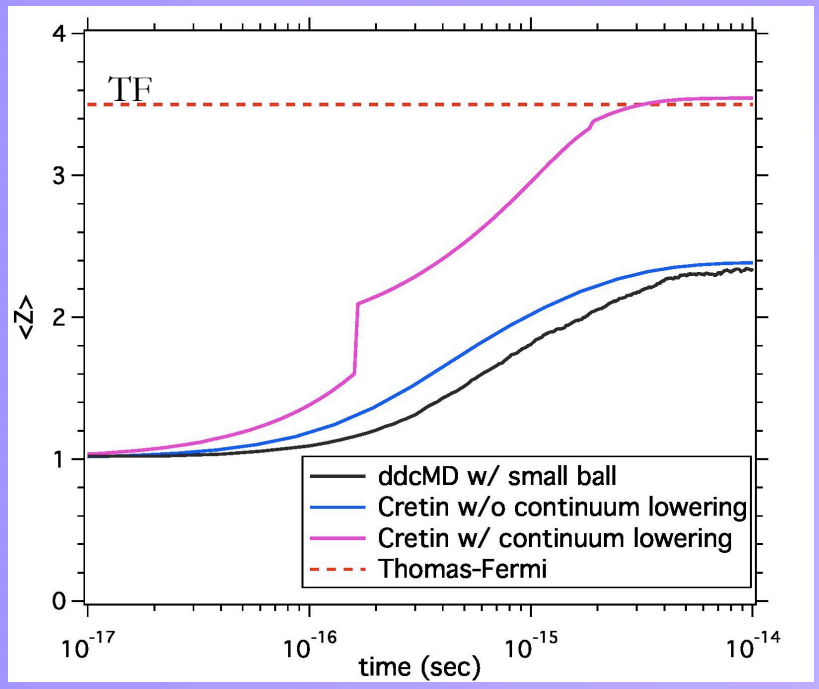
Cretin—a radiative transfer capability for laboratory plasmas

H.A. Scott,
Journal of Quantitative Spectroscopy & Radiative Transfer 71 (2001) 689–701

Classical MD predicts the wrong eigenspectrum –
No gap between cores and free electrons

Warm Dense Carbon Results
Monte Carlo atomic physics

First test of the Monte Carlo core dynamics: approach to equilibrium
Start from near-neutral atoms, use free atom core levels



ddcMD <Z> lacks continuum lowering in this simulation

Monte Carlo Kinetics and equilibrium are consistent with models

1) Quantum Mechanics Via Improved Statistical Potentials

We have attractive e-i forces, but cannot have a 1/r potential for r->0!

Method of Statistical Potentials (SPs):

1. Begin with *exact* QM result for equilibrium behavior
   (i.e., density correlation functions, thermodynamics/EOS)
2. Derive a classical potential to duplicate the QM result:
   - ✓ Now the QM partition function can be sampled using classical molecular dynamics algorithms
3. Finally, we assume that the equilibrium sampling can be used in a real dynamics
   - ✓ long-range classical part still "exact"
   - ✓ the QM "corrections" only truly accurate near equilibrium (structure/screening)

Consider the (exact) partition function in the grand canonical ensemble:

$$\Xi = \sum_N z^N Q_N$$

$$Q_N = Tr\left[\hat{\rho}_N\right] = Tr\left[\exp\left(-\beta \hat{H}\right)\right]$$

$$\Xi \equiv \sum_N z^N \frac{1}{N!\Lambda^{3N}} \int d^3 r_1 \cdots d^3 r_N W(r_1, \ldots r_N)$$ *looks classical*

$$\frac{2\pi\hbar^2}{mT}$$

We can write this, suggestively, as:

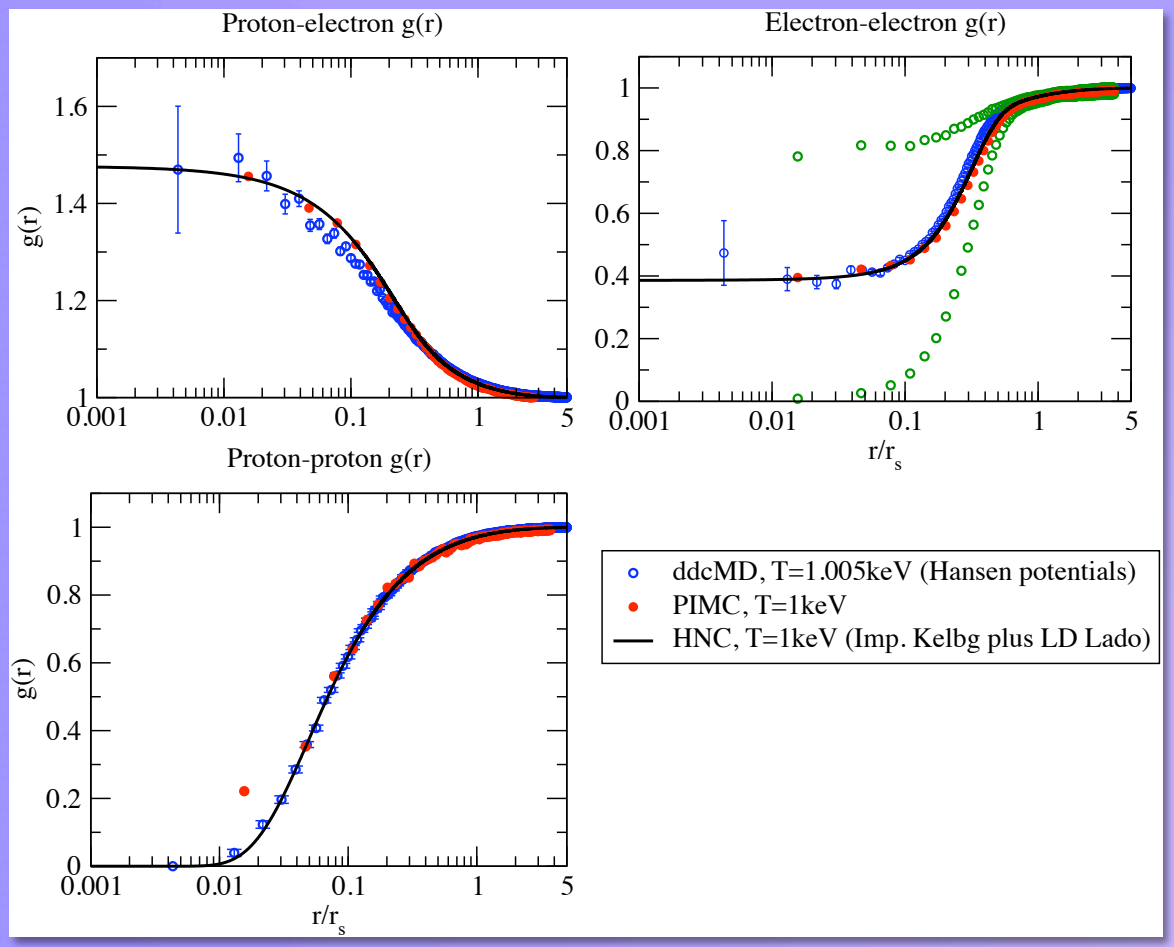
$$W(r_1, \ldots r_N) \equiv \exp\left(-\beta U\left(r_1, \ldots r_N\right)\right)$$ *still exact*

$$\approx \exp\left(-\beta \sum_{i<j} u_{ij}\left(r_{ij}\right)\right)$$ *pair approximation*

- *manipulate QM density matrix to have this functional form*
- *check accuracy with comparison with PIMC*

Existing potentials treat equilibrium correlations in hydrogen

High density, temperature, weak coupling

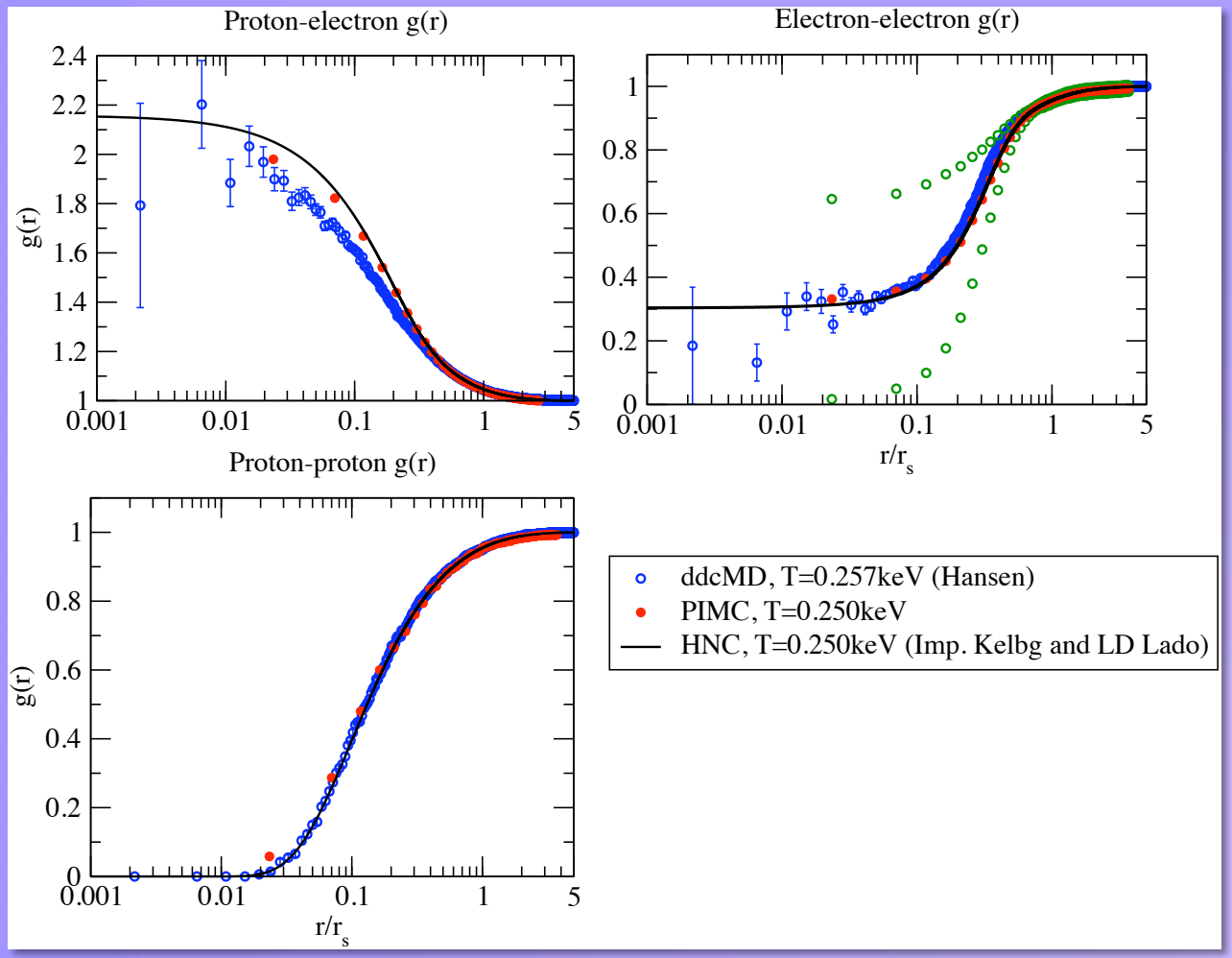


$n_p=n_e=1e25/cc$
$T=1keV$

Difference between spin-parallel, -antiparallel in green -> Pauli term

Existing potentials treat equilibrium correlations in hydrogen

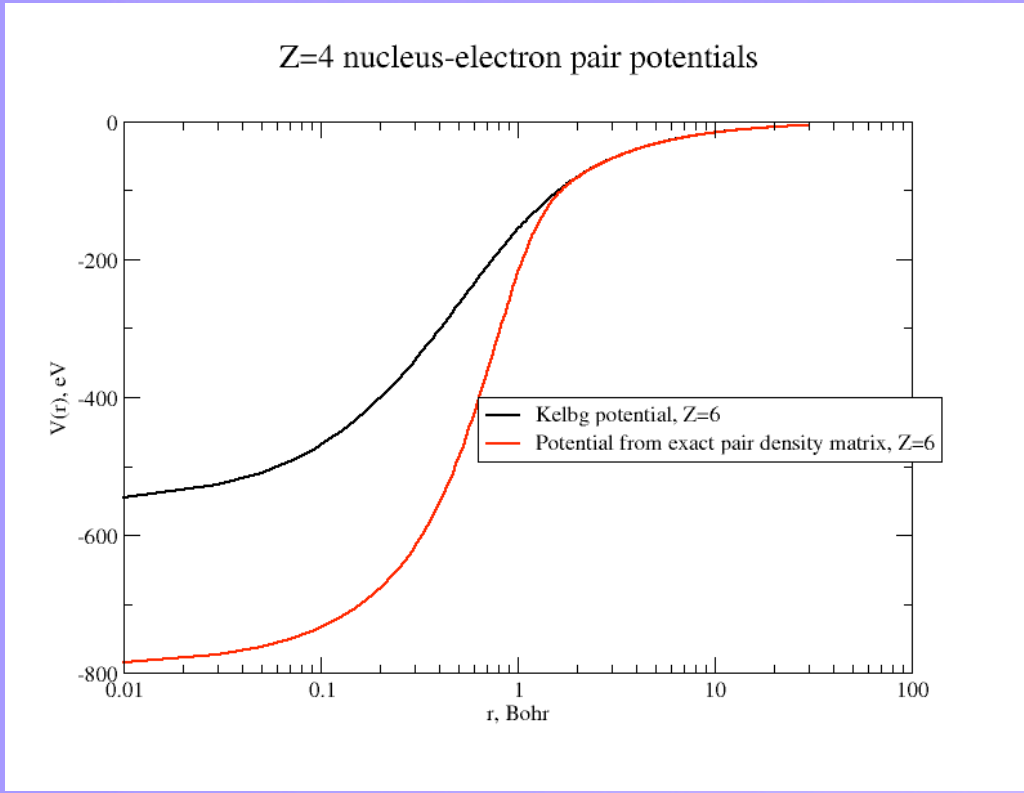
Slightly stronger coupling



$n_p=n_e=1.25e24/cc$
T=0.250keV

We are deriving improved potentials from fully quantum path integral Monte Carlo calculations

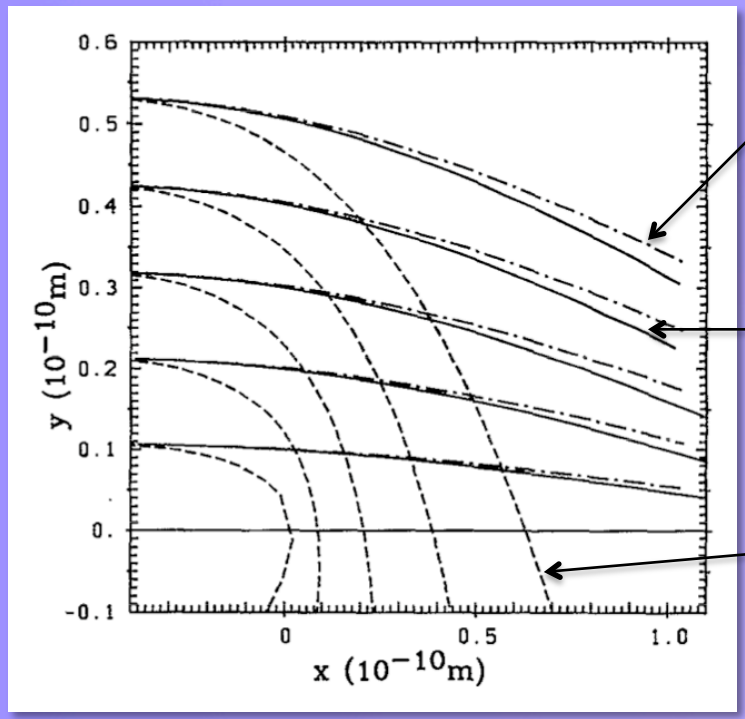
New potentials correctly account for the density from quantum bound states, by design



We are in the process of integrating these potentials into large scale MD

What should the effective scattering cross sections be?

Trajectories for Coulomb (e-p) scattering:



time-dependent variational principle (i.e., WPMD)

"exact" (i.e., numerical solution of Schrödinger equation)

classical

Semiclassical molecular dynamics for strongly coupled Coulomb systems

D. Klakow, C. Toepffer, and P.-G. Reinhard
Institut für Theoretische Physik II, Staudtstr. 7, Universität Erlangen-Nürnberg, D-91058 Erlangen, Germany

What do we expect?

Indeed, QM mainly weakens the Coulomb interaction at short range.

We have confirmed these results using different numerical methods (MPSoft), and different formalisms (Wigner trajectories)

N.B.: This is just Ehrenfest's theorem in action:

$$m\frac{d^2\langle\hat{r}\rangle}{dt^2} = -\langle\nabla U(\hat{r})\rangle$$
$$= -\int d^3r\,|\psi(r,t)|^2\nabla U(r)$$
$$\neq -\nabla U(\langle\hat{r}\rangle)$$

Centroid of QM trajectory does not follow CM trajectory

We must capture this short range behavior

Scattering is being compared to full quantum simulations

MP/SOFT method

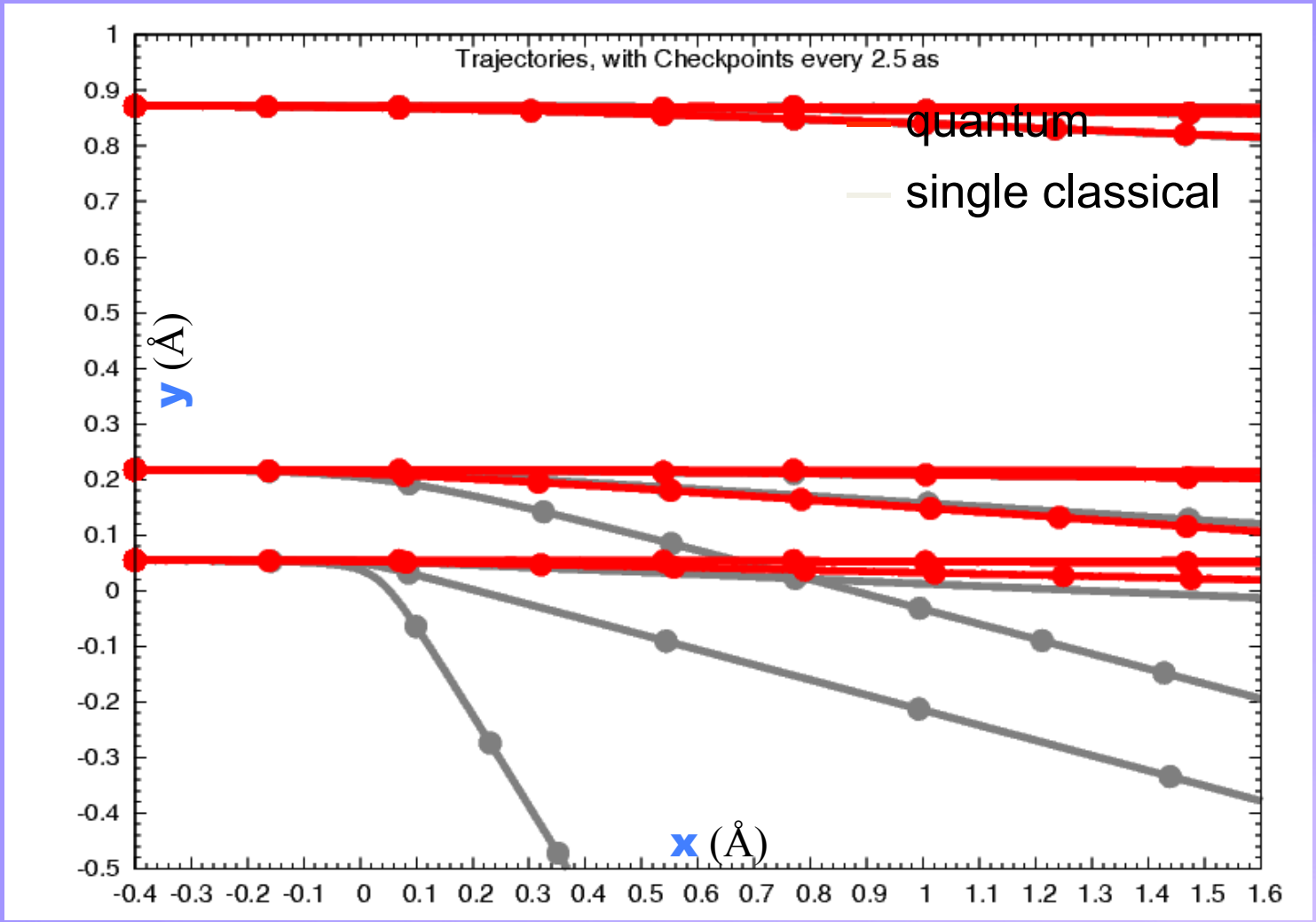
Wu,Y.; Batista, V.S. *J. Chem. Phys.* (2003) **118**, 6720
Wu,Y.; Batista, V.S. *J. Chem. Phys.* (2003) **119**, 7606
Wu,Y.; Batista, V.S. *J. Chem. Phys.* (2004) **121**, 1676
Chen, X., Wu,Y.; Batista, V.S. *J. Chem. Phys.* (2005) **122**, 64102
Wu,Y.; Herman, M.F.; Batista, V.S. *J. Chem. Phys.* (2005) **122**, 114114
Wu,Y.; Batista, V.S. *J. Chem. Phys.* (2006) **124**, 224305
Chen, X.; Batista, V.S. *J. Chem. Phys.* (2006) **125**, 124313
Chen, X.; Batista, V.S. *J. Photochem. Photobiol.* (2007) **190**, 274
Kim, J.; Wu, Y.; Batista, V.S. *Israel J. Chem.* (2009) **submitted.**
Chen, X.; Batista, V.S. *J. Chem. Phys.* (2009) **in prep.**

Trotter expansion:

$$e^{-(i/\hbar)\hat{H}\tau} \approx e^{-(i/\hbar)\hat{V}\tau/2}\mathrm{FT}^{-1}\, e^{-(i/\hbar)\frac{\mathbf{p}^2}{2m}\tau}\mathrm{FT}\, e^{-(i/\hbar)\hat{V}\tau/2}.$$

Here, $\hat{H} = \frac{\mathbf{p}^2}{2m} + \hat{V}(\mathbf{x})$, and FT indicates the action of the multidimensional Fourier transform.

SINGLE CLASSICAL TRAJECTORIES

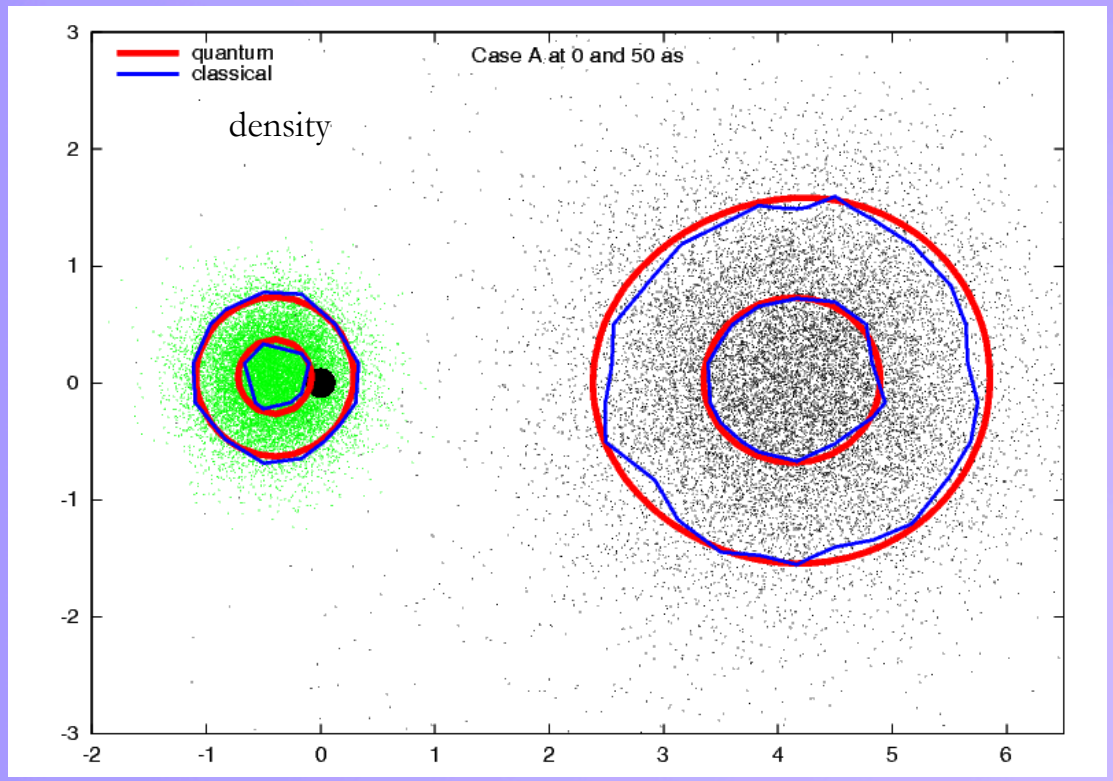


- No problems with high initial momenta, velocities are correct.
- Single classical particles qualitatively different at small impact params.

Going beyond expectation values,
Classical behavior can be compared to quantum distributions

Quantum distribution is modeled by a cloud of particles with initial density $|\Psi|^2$
e.g., using 15625 particles:

INITIAL KE=0.5keV IMPACT PARAMETER=0.0545 Å



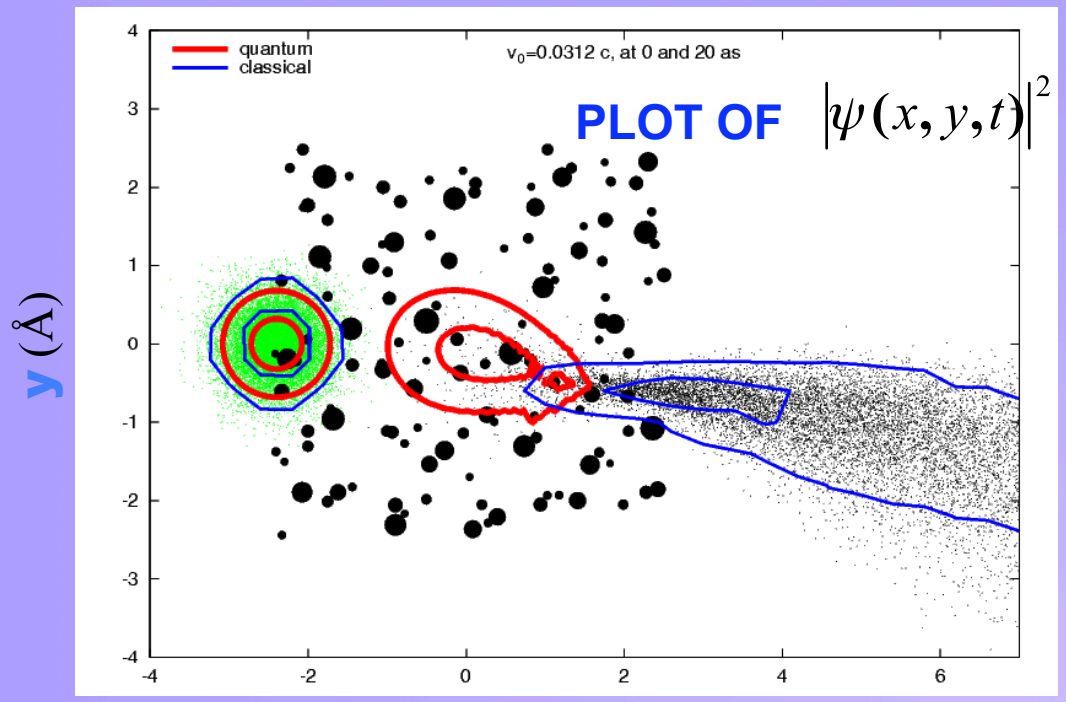
y (Å)

x (Å)

Coherent multiple scattering can alter propagation

Cubic element of 125 protons liquid structure, fixed coordinates, zero temperature
Electron E_k = 0.5keV, initial impact parameter = 0Å

Classical density moves ahead of quantum density.



y (Å)

x (Å)

We also have wave packet MD methods to apply.

Path so far

1. Directly applied classical MD to model problems, "exact" numerical calculations

2. Used existing quantum corrected potentials to avoid the classical Coulomb catastrophe
   apply to opposite-charged problems, neutral plasmas, e.g. C (or H)

3. Conducted initial stopping experiments

4. Developed improved quantum atomic dynamics
   cross sections, detailed balance, equilibria

5. Developed advanced quantum potentials for near equilibrium conditions

…

6. Integrate corrections into overall MD, assess accuracy