Determining dE/dx in Warm Dense Matter Using Non-Equilibrium Molecular Dynamics

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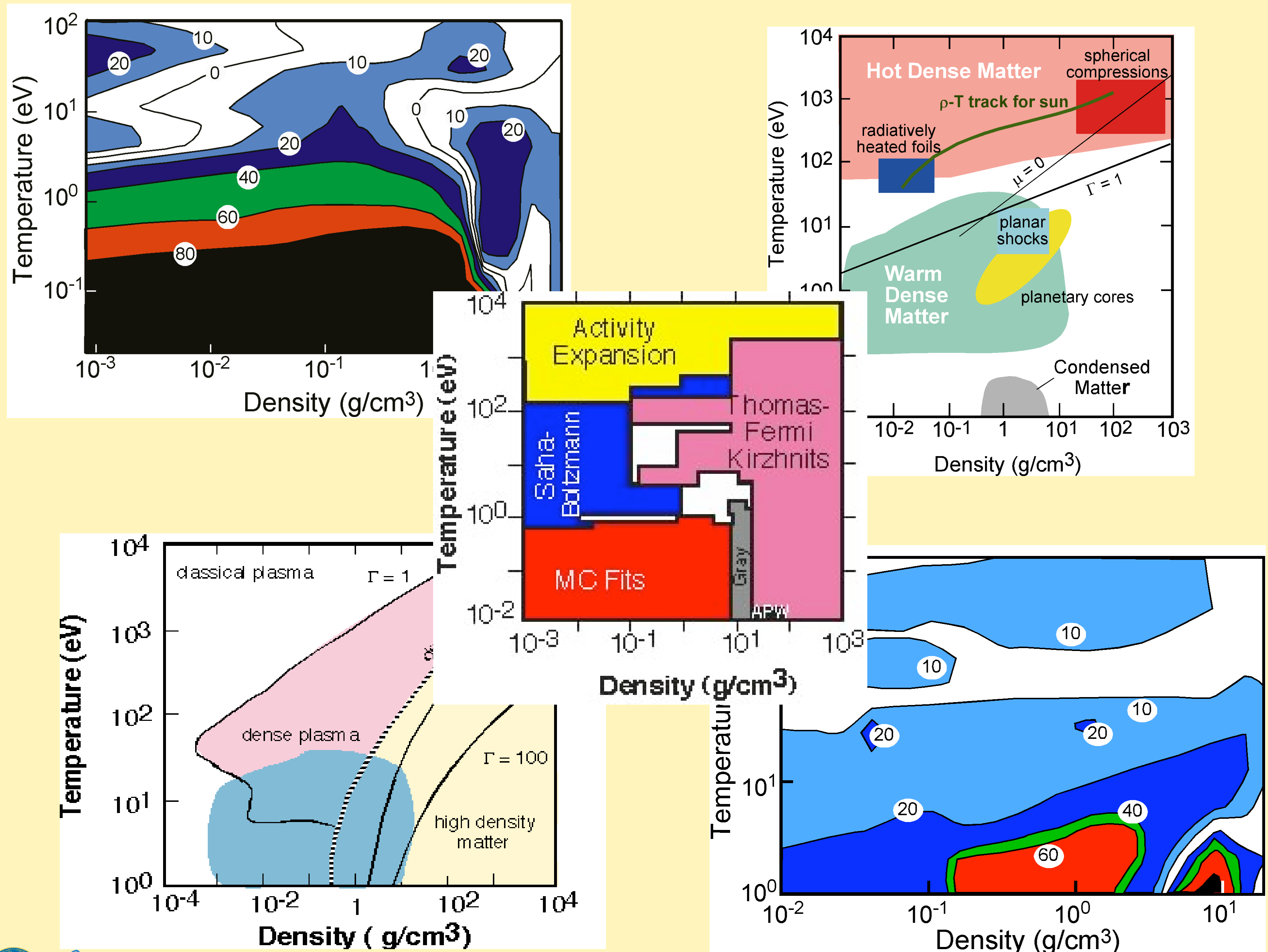
Outline:

- Main goals of this work
- Our analytic model
- Our molecular dynamics model and method
- Issues and results

*On leave from: Theoretical Division, Los Alamos National Laboratory



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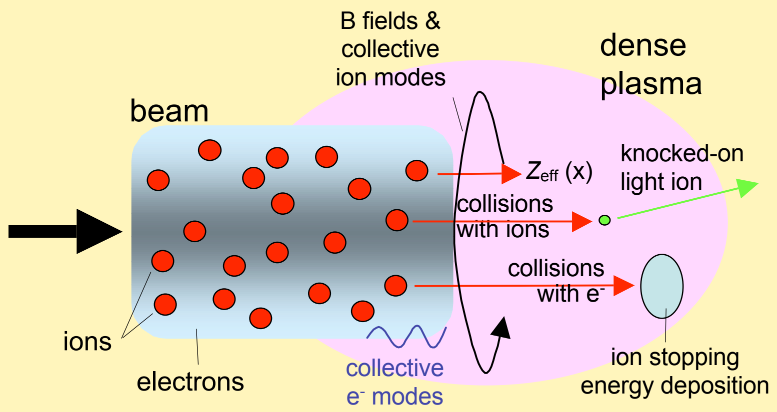
Main Goals of This Work

The obvious three:

- How do particles stop in WDM?
- How can we create WDM with stopping particles?
- How can we diagnose WDM with projectiles?



More specifically:

✓ What is different in WDM, relative to gases, cold solids, and ideal plasmas?

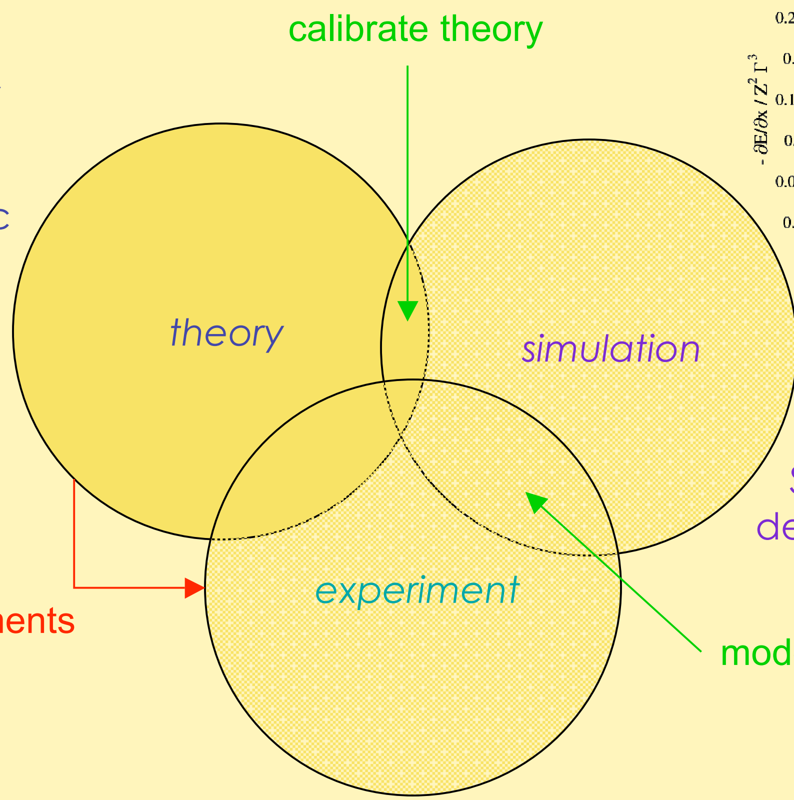
- partial degeneracy (Pauli blocking)
- strong coupling within target
- atomic physics within target (continuum lowering, incipient Rydberg states)
- radiation
- strong projectile-target interaction (resonant capture)
- Etc.

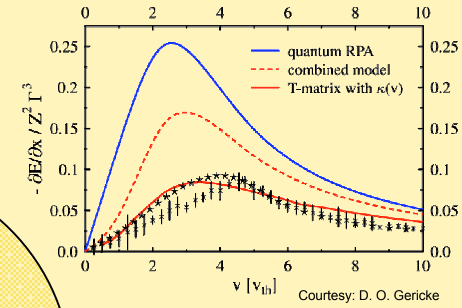
✓ What "analytic" models can we construct for experimental design purposes?

✓ How can accuracy and self-consistency be quantified with simulation?

We Combine Analytic and Simulation Capabilities

Theory is highly developed, but has weaknesses in treating everything (nonlinear response, atomic physics, collective excitation, etc.) self-consistently.





Simulation is much less developed, but has many strengths.

For WDM, what type of simulation is needed?

- fully dynamic electron & ion responses (nonequilibrium excitation)
- strong projectile-target scattering (accurate trajectories)
- strong coupling in target (discrete particle information)
- partial degeneracy of target (Pauli over wide range of temperature)
- nonlinear screening of projectile by target (electron trapping, bound states)

Our Current Analytic Model

$$\frac{dE}{dx} = \frac{e^2}{\pi v^2} \int_0^\infty \frac{dk}{k} \left| Z - n_b(k) \right|^2 \int_{-kv}^{kv} d\omega \; \omega \, \mathrm{Im}\left[\frac{1}{\varepsilon(k,\omega)} \right] n_B(\omega)$$

effective charge

We decompose the dielectric response function as:

$$\frac{1}{\varepsilon(k,\omega)} = 1 + v(k) \frac{\chi^{(0)}(k,\omega)}{1 - v(k)\chi^{(0)}(k,\omega)\left[1 - G(k,\omega)\right]}$$

plasmon excitation

The free-particle response is given by the finite-temperature Lindhard function:

$$\chi^{(0)}(k,\omega) = 2\sum_q \frac{f(q) - f(k+q)}{\hbar\omega - \left(\varepsilon_{k+q} - \varepsilon_q\right) + i\delta}$$

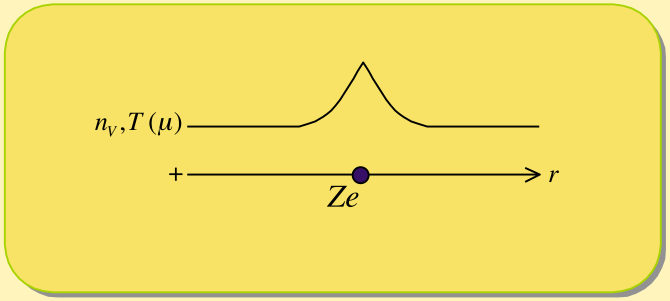
free-particle density fluctuations, including Pauli blocking and diffraction

Various forms for the dynamic local field correction are known, but we neglect them for this talk.

$$G(k,\omega) = 0$$

strong coupling

Effective Charge: Drifting, Modified Thomas-Fermi Model



Assumptions:

- projectile is "slow"
- charge renormalization is the dominant nonlinear interaction
- Thomas-Fermi is a reasonable starting point
- quantum (gradient) correction included via pseudopotential

Consider a drifting Fermi-Dirac:

$$n(r) = 2\int \frac{d^3p}{(2\pi\hbar)^3} \frac{1}{\exp\left[\beta\left(\left(\vec{p}+m\vec{v}\right)^2/2m + u(r) - \mu\right)\right]+1}$$

Pseudopotential chosen to be of the form:

$$u(r) = -\frac{Ze^2}{\sqrt{r^2+a^2}}\exp\left(-r/\lambda_{TF}(\mu)\right)$$

Parameter *a* constrained by the condition:

$$Z = \int d^3r\left[n(r) - n(\infty)\right]$$

Separate bound and free contributions:

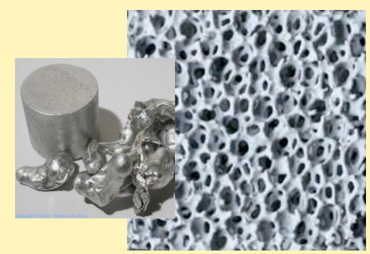
$$n_b(r):\ \left(\vec{p}+m\vec{v}\right)^2/2m + u(r) < 0$$

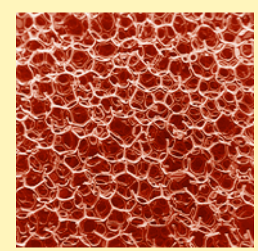
$$n_f(r):\ \left(\vec{p}+m\vec{v}\right)^2/2m + u(r) > 0$$

The effective projectile is the nucleus and its bound electrons.

This model has:

- arbitrary target density and temperature
- finite density at nucleus
- exact linear result
- perfect screening
- velocity-dependent charge
- finite-size bound cloud (effective charge)





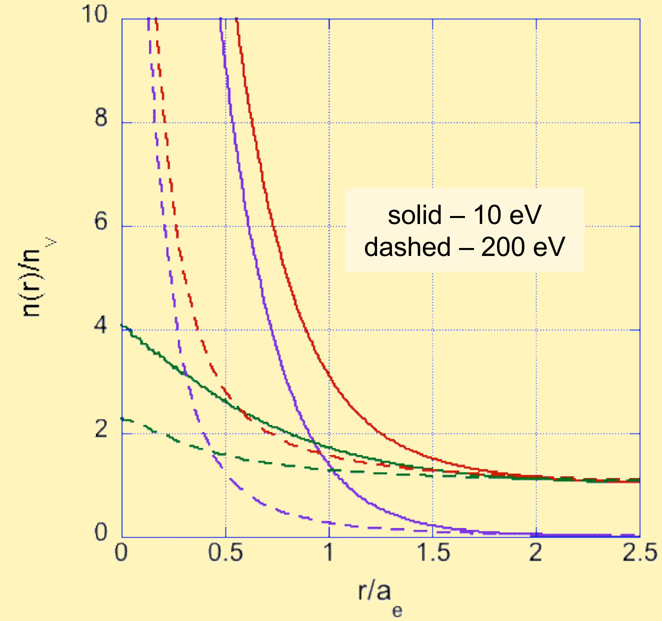
Examples of Effective Charge Calculations

Consider a *Na* ion stopping in *Al*:

Density profiles at zero velocity:

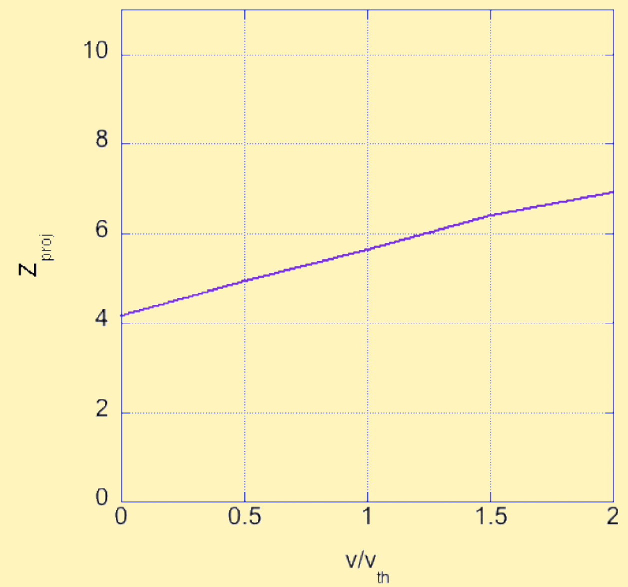
$$Z = 4.2\ (T = 10eV)$$
$$Z = 8.5\ (T = 200eV)$$

~4x in stopping

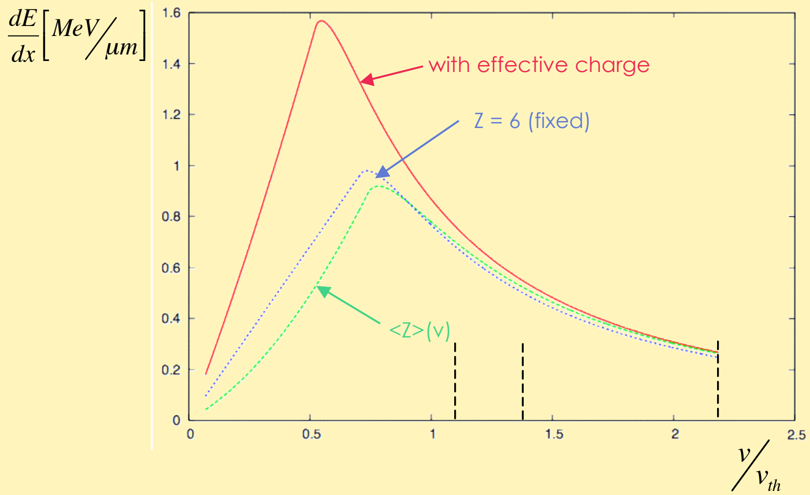


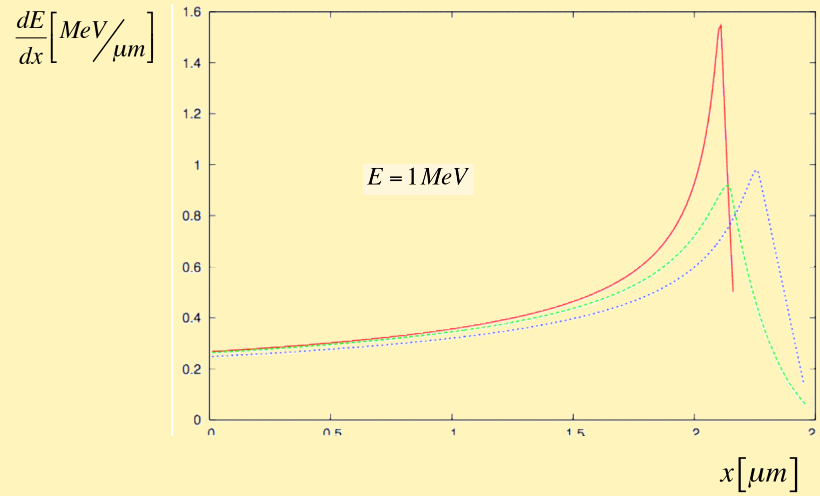
N.B.: Free electron screening is weak.

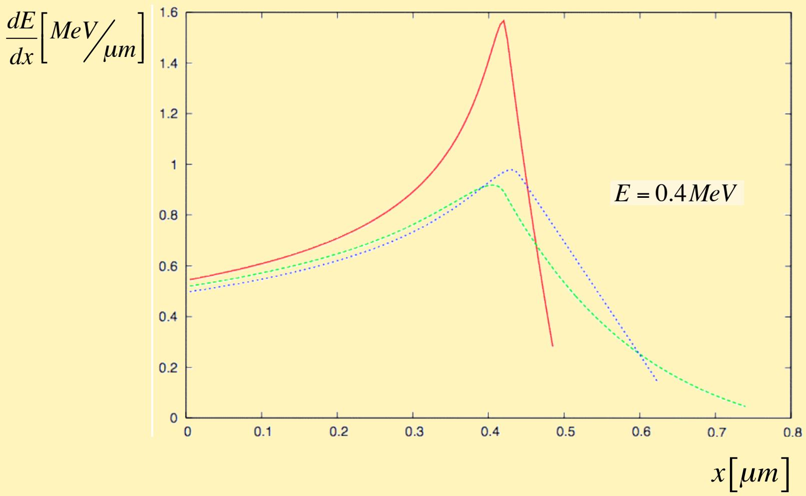
Charge state versus velocity:
(T=10eV)



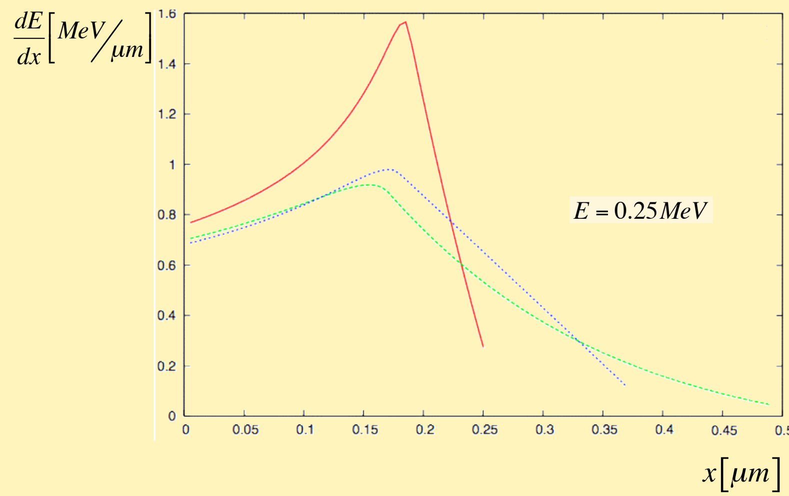
Analytic Model Results for Na Projectile in Al at T=10eV







Note change of scale.



Note change of scale.

We Use Molecular Dynamics Methods

Molecular dynamics means:
• Solve the equations of motion exactly

Molecular dynamics does NOT:
• use a mesh - detailed trajectories are followed
• use the Born-Oppenheimer approximation – electrons are dynamic
• assume equilibrium distributions – applicable to nonequilibrium

This comes with a price:
• few particles (N~thousands) – use periodic boundary conditions
• forces tend to be classical-like – use effective quantal interactions
• statistical “noise” can be large – use several ensembles

We Obtain Quantal Interactions from Partition Function

Consider the partition function of a quantum system:

$$\begin{aligned}\exp(-\beta F) &= \mathrm{Tr}[\hat{\rho}] = \mathrm{Tr}\left[\exp\left(-\beta \hat{H}\right)\right] \\ &= \int d^3r_1 \ldots d^3r_N \langle r_1,\ldots,r_N | \exp\left(-\beta \hat{H}\right) | r_1,\ldots,r_N \rangle \\ &= \int d^3r_1 \ldots d^3r_N \, \mathsf{F}(r_1,\ldots,r_N) \\ &\cong \int d^3r_1 \ldots d^3r_N \, \mathsf{G}(r_{12}, r_{13}\ldots, r_{N-1,N}) \\ &\equiv C\int d^3r_1 \ldots d^3r_N \exp\left(-\beta \sum_{i<j} u_{ij}(r_{ij})\right)\end{aligned}$$

Currently, we use:

diffractive scattering — spin-averaged Pauli exclusion

$$u_{ab}(r,\lambda\!\!\!^{-}_{ab}) = -\frac{Z_a Z_b e^2}{r}\left(1 - e^{-r/\lambda\!\!\!^{-}_{ab}}\right) + \delta_{ae}\delta_{be} T \ln(2)\, e^{-r^2/\pi \ln(2) \lambda\!\!\!^{-2}_{ab}}$$

thermal deBroglie wavelength

Some exact limits:

- classical, strongly-coupled plasma
- ideal Fermi gas pair correlation function for zero separation: $g_0(0)=0.5$

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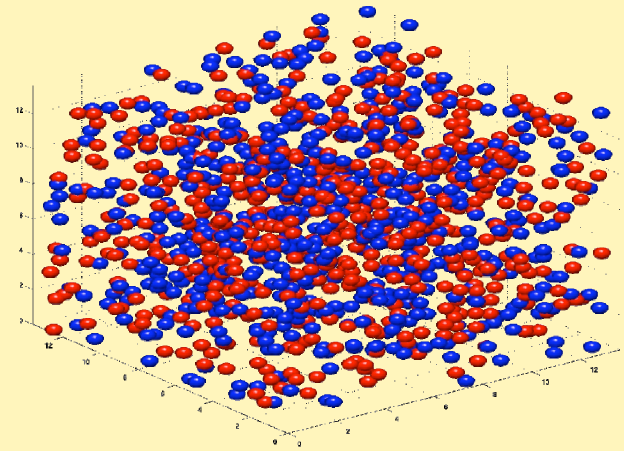
Some Details

Our current MD capability is:
- electrons *and ions* (quasi-bound states, knock-ons, energy split)
- projectile
- several thousand particles



Newton's equations for N particles are solved via velocity-Verlet:

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

$$\vec{v}(t+\Delta t) = \vec{v}(t) + \frac{1}{2}\left(\vec{a}(t+\Delta t) + \vec{a}(t)\right)\Delta t$$

- establish initial equilibrium via equilibration phase (~20,000 steps) "data" accumulated with no thermostat
- inject projectile
- typical time step ~$0.02/\omega_{pe}$

The forces include pure Coulomb, diffractive, and Pauli terms:

$$H = \sum_a \frac{p_a^2}{2m_a} + \sum_{a<b}\left[\frac{q_a q_b}{r_{ab}}\left(f(\alpha, r_{ab}) - \exp\left(-r_{ab}/\lambda_{ab}\right)\right) + g(\alpha, r_{ab}) + T_e \ln(2)\exp\left(-r_{ab}^2/\pi\ln(2)\lambda_{ee}^2\right)\right]$$

$$\lambda_{ab} = \frac{\hbar}{\sqrt{\pi\mu_{ab}T}}$$

Shape of main cell minimized via spherically-averaged Ewald:

$$f(\alpha, r_{ab}) = \mathrm{erfc}(\alpha r_{ab})$$

$$g(\alpha, r_{ab}) = \frac{4\pi}{L^3}\sum_{\vec{k}\neq\vec{0}} \frac{\exp(-k^2/4\alpha^2)\sin(kr_{ab})}{k^3 r_{ab}}$$

- $N_{max}=10$
- $g(\alpha,r)$ tabulated across 500 bins
- energies and forces tabulated separately
- 2nd-order Newton-Gregory interpolation
- other forces/energies computed directly

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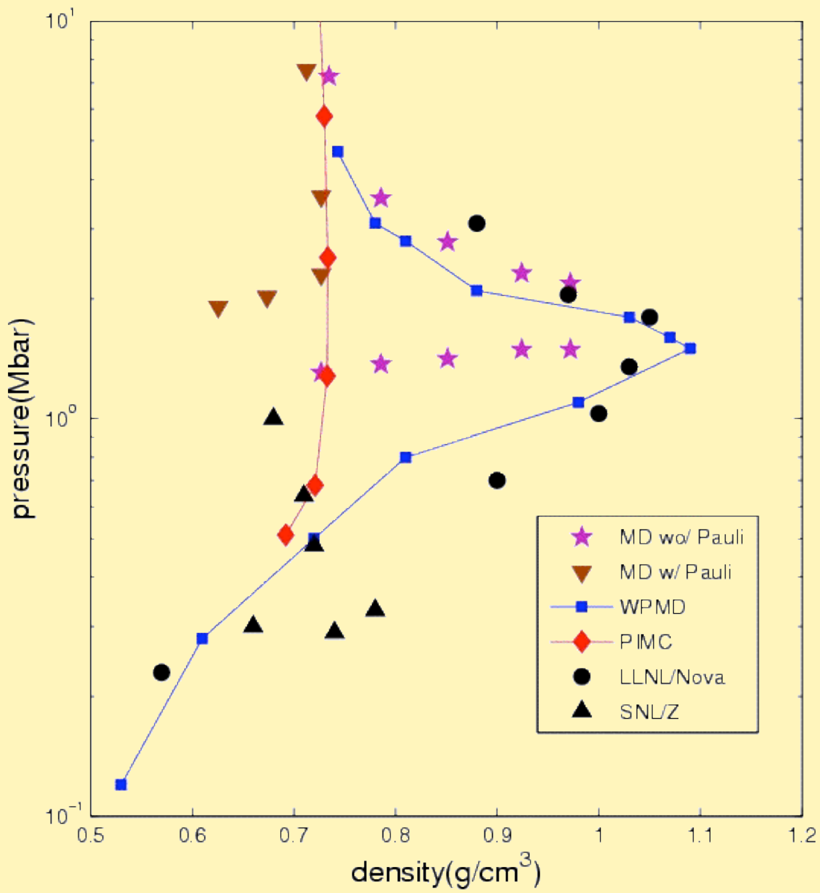
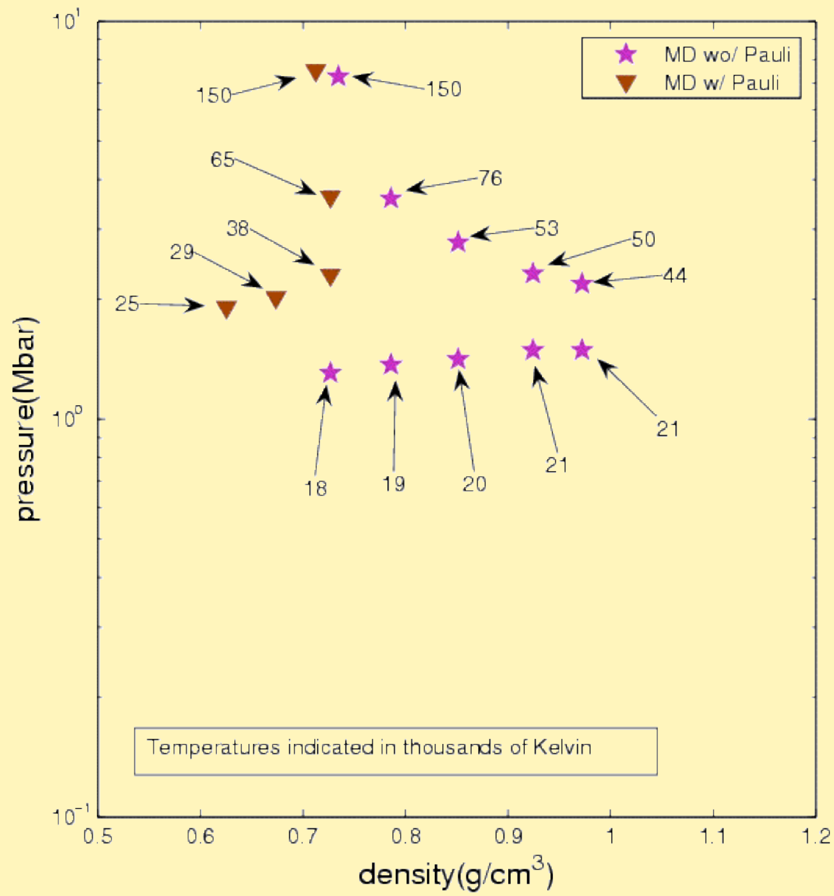
Our MD Physics Model Agrees Well with Experiment

Deuterium Hugoniot



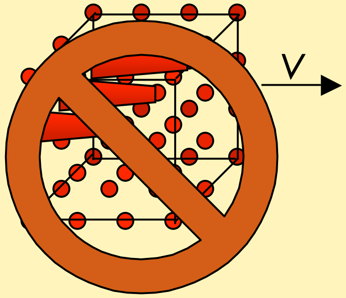
comparison with other models
and experimental data



corresponding temperatures

Various Issues Arise for Computing Stopping Power

- Need enormous system, not periodic boundary conditions (PBCs)
  - ✓ plasma is *not* periodic on few-particle length scale
  - ✓ beam is *not* a simple-cubic lattice
  - ✓ main cell cannot be big enough to actually stop the projectile



- Need to resolve wake potential*
  - ✓ PBCs yield wake-wake, wakes-projectile interaction
  - ✓ contributions to stopping arise from very long wavelengths (hydro-scale)

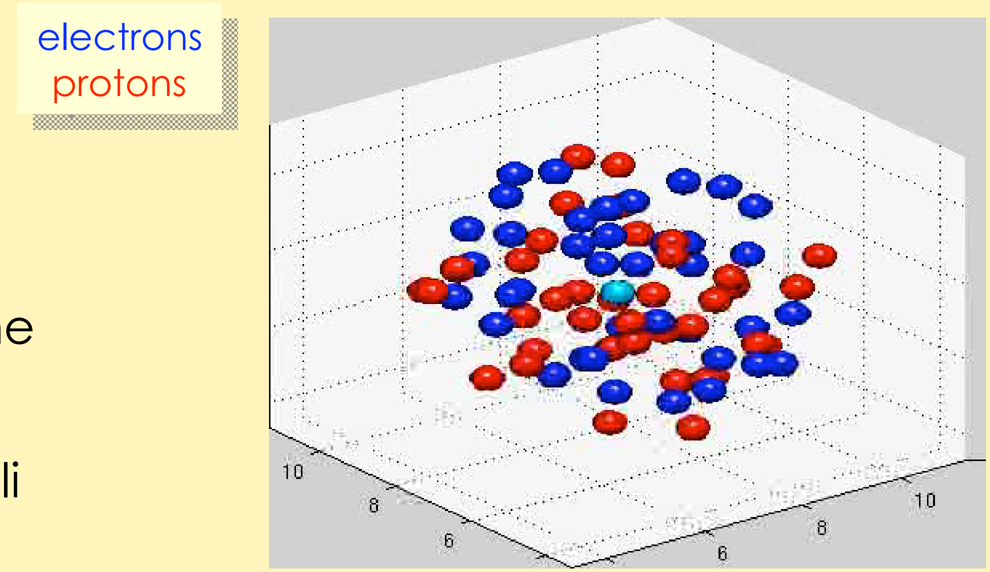
- Need to obtain steady-state response
  - ✓ inserting a projectile for each *v*, *Z* unphysically "shocks" the plasma
- Need accurate plasma physics
  - ✓ target is initially partially degenerate and strongly coupled

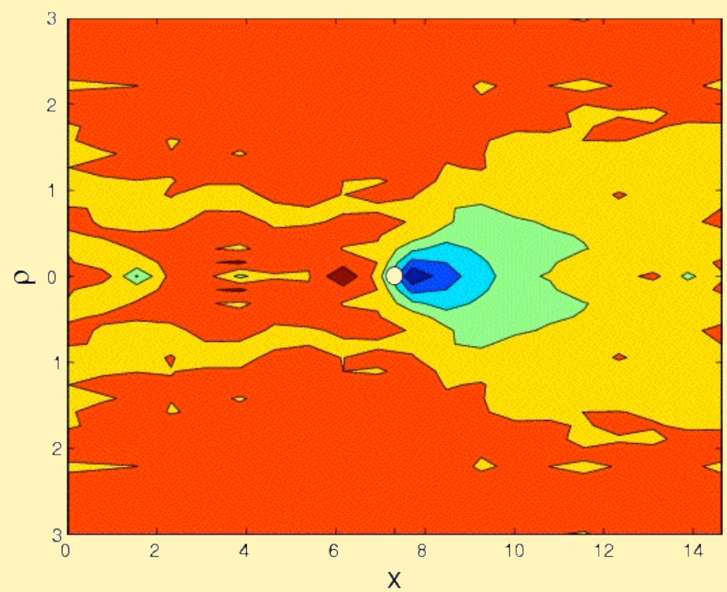
- Need accurate atomic physics
  - ✓ charge state can change by many (micro-scale)
- Need to resolve various time scales
  - ✓ transients, collective modes, electrons/ions, bound electrons

Wake Size/Shape and Periodic Boundary Conditions

Simulation example:

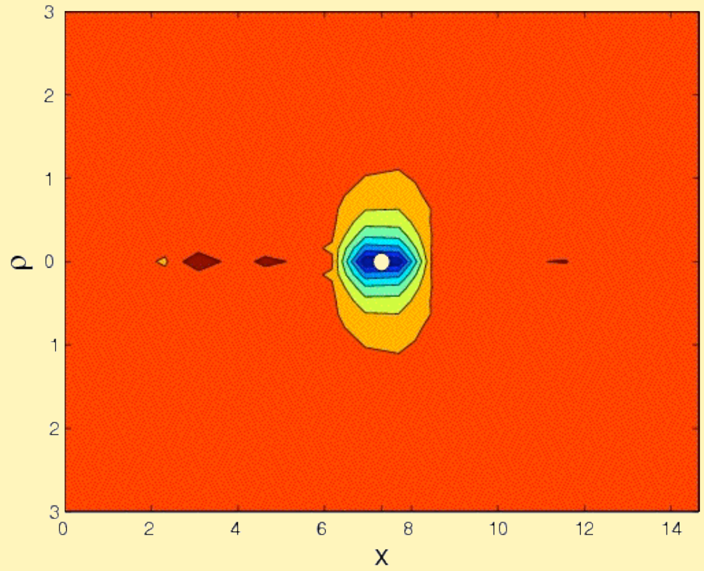
- in projectile reference frame
- N=1,501 (750e, 750p, 1proj)
- Deutsch diffractive, no Pauli
- Standard Ewald
- M/m = 10
- $n_{e,p}=10^{24}$ cm^{-3}
- $T_{e,p}$=100 eV
- Z_{proj}=+30
- E_{proj}=1 MeV



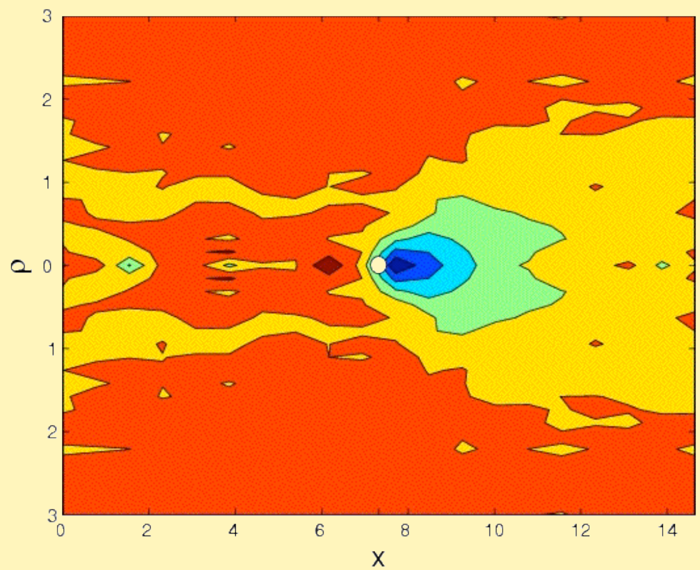


PBCs Dominate for Fast Projectiles

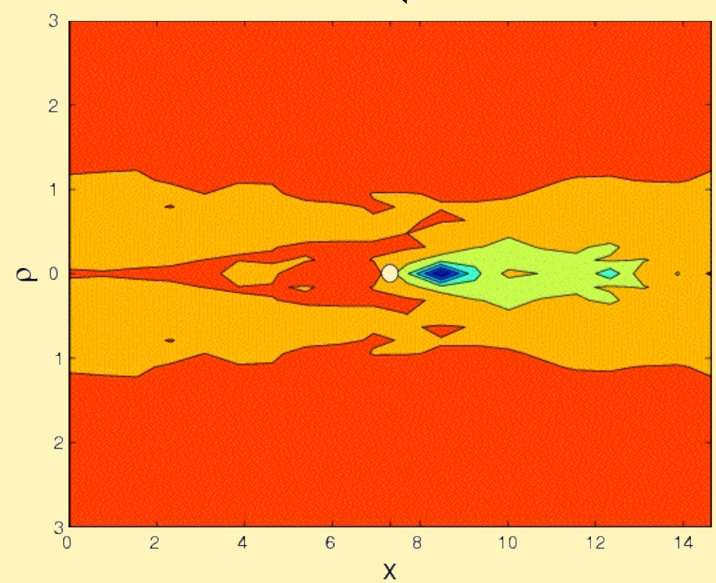
Projectile at rest (“DH screening”)



Moderate velocity (1 MeV/u)



High Velocity (10 MeV/u)



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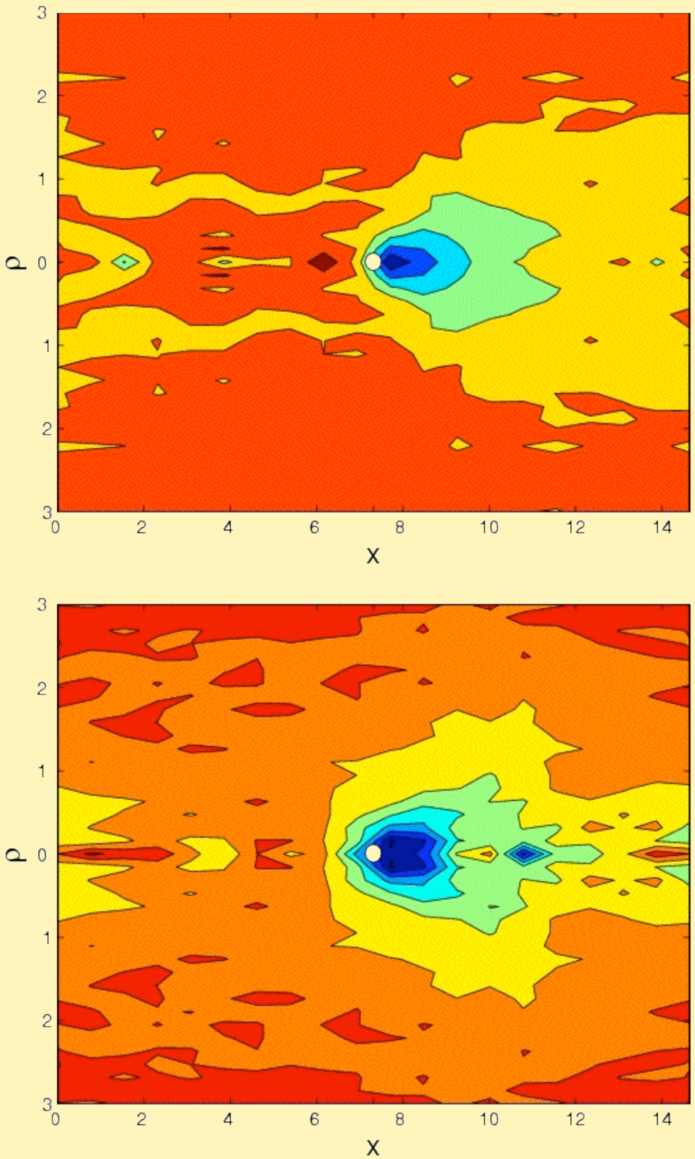
Upstream Re-Thermalization Helps

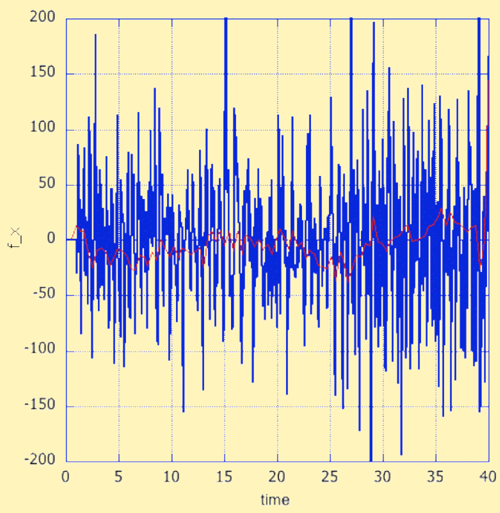
Simulation example:

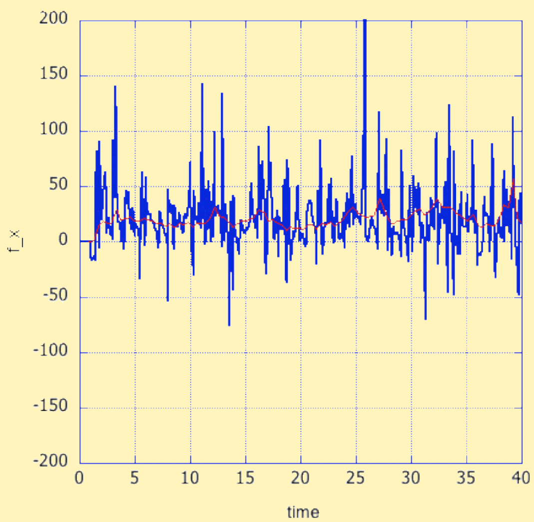
- in projectile reference frame
- N=1,501 (750e, 750p, 1proj)
- Deutsch diffractive, no Pauli
- Standard Ewald
- M/m = 10
- $n_{e,p}=10^{24}$ cm^{-3}
- $T_{e,p}$=100 eV
- Z_{proj}=+30
- E_{proj}=1 MeV
- plasma velocity randomized ahead of projectile

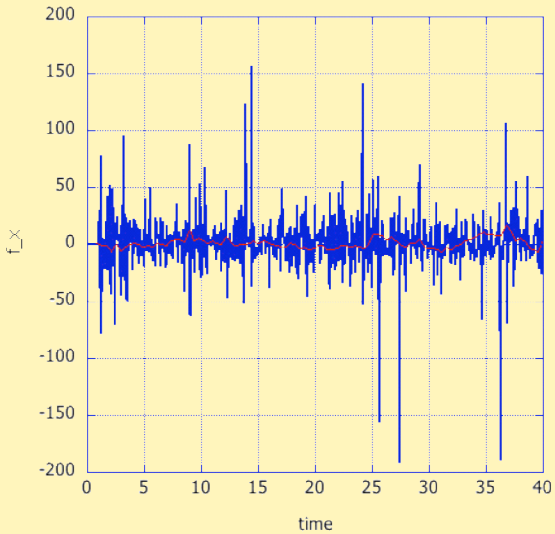


Stopping Occurs From Average Force On Projectile

Molecular dynamics gives the force directly on the projectile.





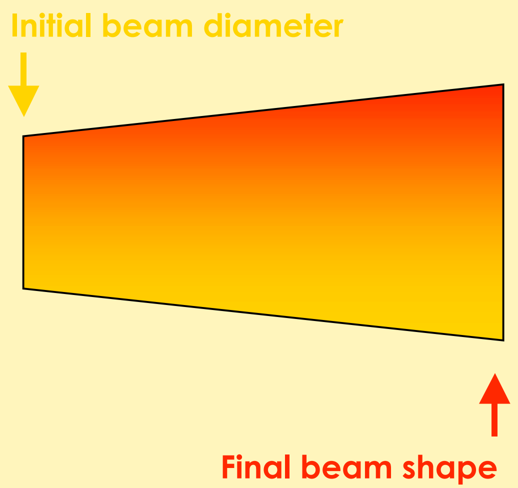


Very slow (E=0). Intermediate velocity (E=1MeV/u). Fast (E=10MeV/u).

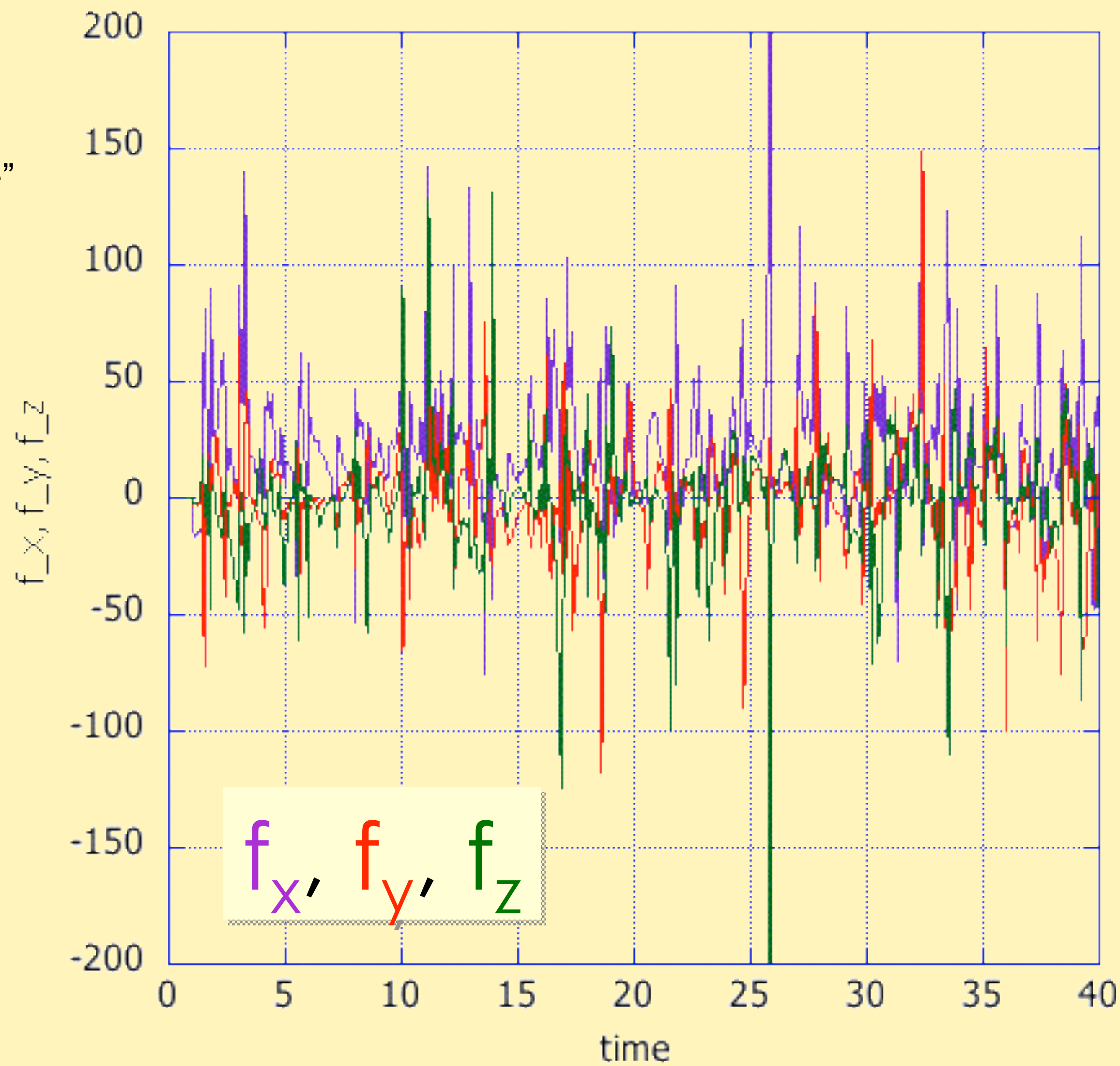
Note that we are looking for a needle in a haystack!

MD Naturally Has All Force Components

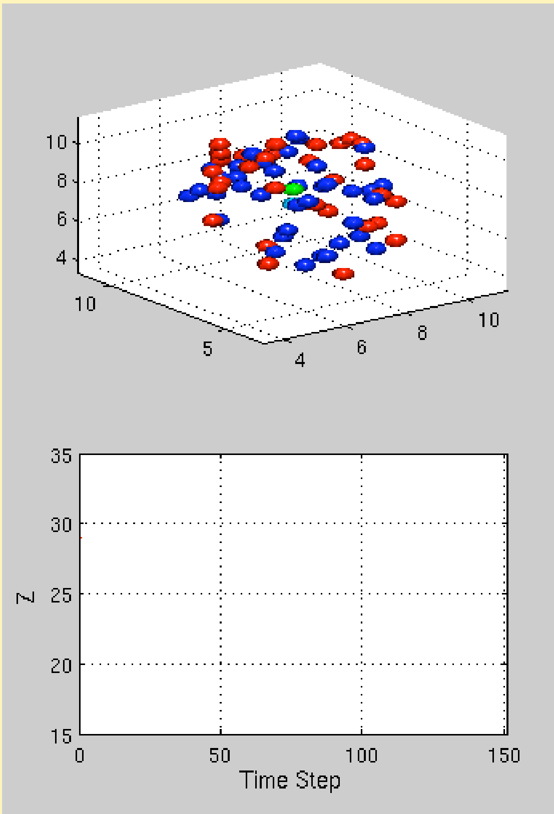
Incident beam does not:
- travel along a line
- deposit energy "on average"



This rapid microfield will affect spectral line emission - diagnostic!

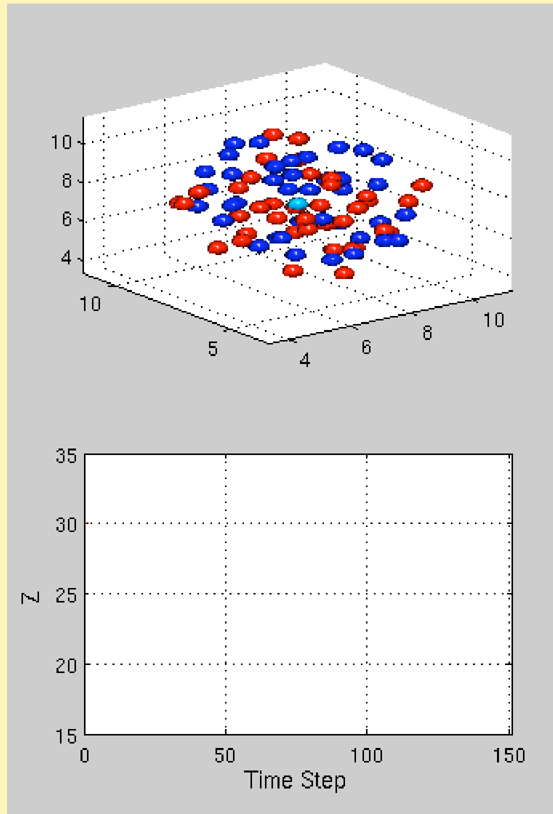


Initial Studies of Effective Charge Underway

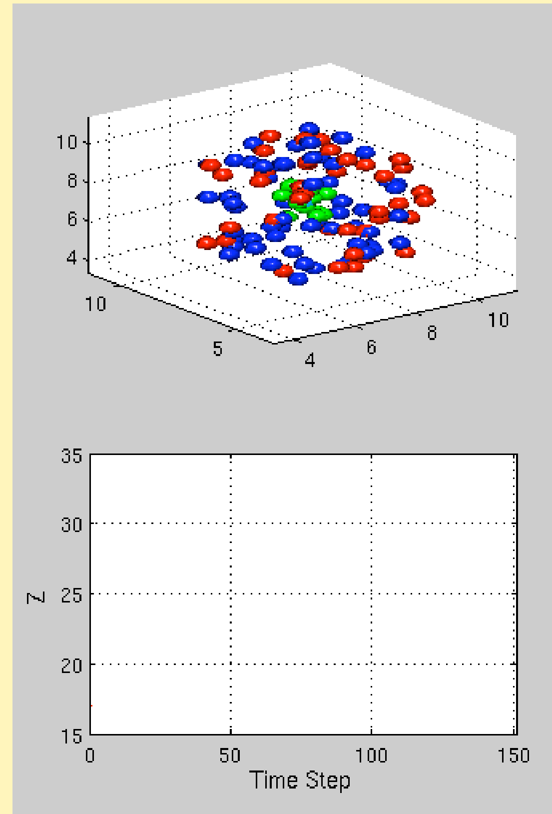


fast

Green spheres represent "bound" electrons.



slow



stopped

Summary

- We have an analytical model for modeling stopping in WDM, and we would like to determine its validity; some theoretical issues can be checked by simulation.

- We have developed a computational tool for studying stopping power
  - physics issues addressed
  - computational issues addressed

- Now, use this tool for specific applications
  - actual stopping-power problems
  - using beams to create plasma experiments (e.g., EOS)
  - calibrate analytic methods in overlap regimes

- Continue to advance physics model
  - spin-resolved, density-dependent Pauli
  - low-temperature diffraction
  - wave-packet molecular dynamics

Welcome to the

Back-Up Slides…

We Are Exploring a Systematic Approach: WPMD

- Time-Dependent Variational Principle

$$\delta \int_{t1}^{t2} dt \left\langle \Psi(\mathbf{z}(t)) \middle| i\hbar \, {}^{d}\!/_{dt} - \hat{H} \middle| \Psi(\mathbf{z}(t)) \right\rangle = 0$$

This is the *time-dependent* Schrödinger equation! (in principle)

- Wave function Ψ is parameterized by functions z(t), whose dynamics are of the form

$$C_{\mu\nu} \frac{dz_\nu}{dt} = \frac{\partial H}{\partial z_\mu}$$

- All physical observables are obtained quantum mechanically:

$$O \equiv \left\langle \Psi \middle| \hat{O} \middle| \Psi \right\rangle$$

- We choose to characterize the wave function as an antisymmetrized product of individual wave packets, e.g. of gaussian or exponential shape

$$\Psi = A\left[\prod_{i,j} \varphi\left(\mathbf{x}_i, \mathbf{z}_j(t)\right)\right]$$

*Feldmeier & Schnack, Rev. Mod. Phys. (2000)

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Example: Wavefunction Evolution for Single Electron

- For example, a Gaussian* WP with parameters $\mathbf{r}(t), \mathbf{p}(t), \gamma(t), p_\gamma(t)$

$$\varphi(\mathbf{x},t) = \frac{1}{(\gamma\sqrt{\pi})^{3/2}} \exp\left[-\left(\frac{1}{2\gamma^2} - i\frac{p_\gamma}{3\hbar\gamma} \right)(\mathbf{x}-\mathbf{r})^2 + i\mathbf{p}\cdot(\mathbf{x}-\mathbf{r})/\hbar \right]$$

  - $\mathbf{r}, \mathbf{p}$ correspond to classical coordinates and momentum, while γ, p_γ provide a quantum width with its canonical momentum

- With these parameters, equations of motion are canonical

$$\frac{d\mathbf{r}}{dt} = \mathbf{p}/m \qquad \frac{d\mathbf{p}}{dt} = -\frac{Ze^2}{r^3}\left[\mathrm{erf}\left(\frac{r}{\gamma}\right) - \frac{2}{\sqrt{\pi}}\frac{r}{\gamma} e^{-r^2/\gamma^2} \right]\mathbf{r}$$

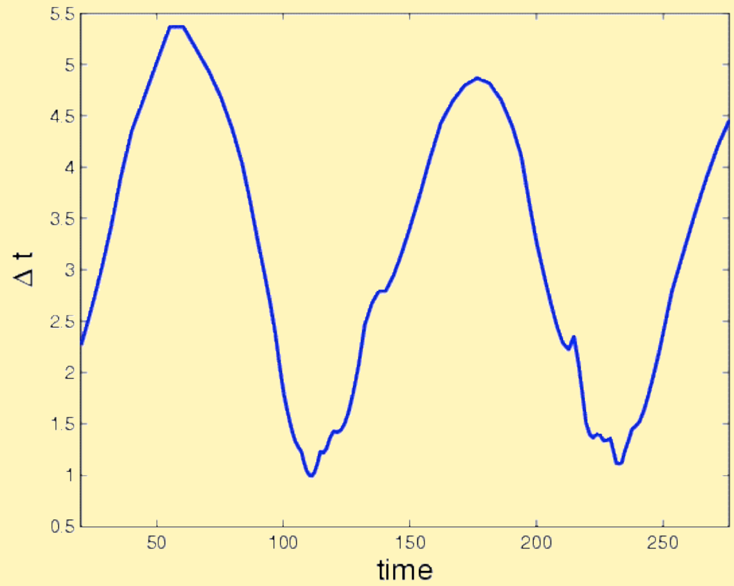
$$\frac{d\gamma}{dt} = 2p_\gamma/3m \qquad \frac{dp_\gamma}{dt} = \frac{3\hbar^2}{2m\gamma^3} - \frac{2}{\sqrt{\pi}}\frac{Ze^2}{\gamma^2} e^{-r^2/\gamma^2}$$

Computational effort ~33% increased.

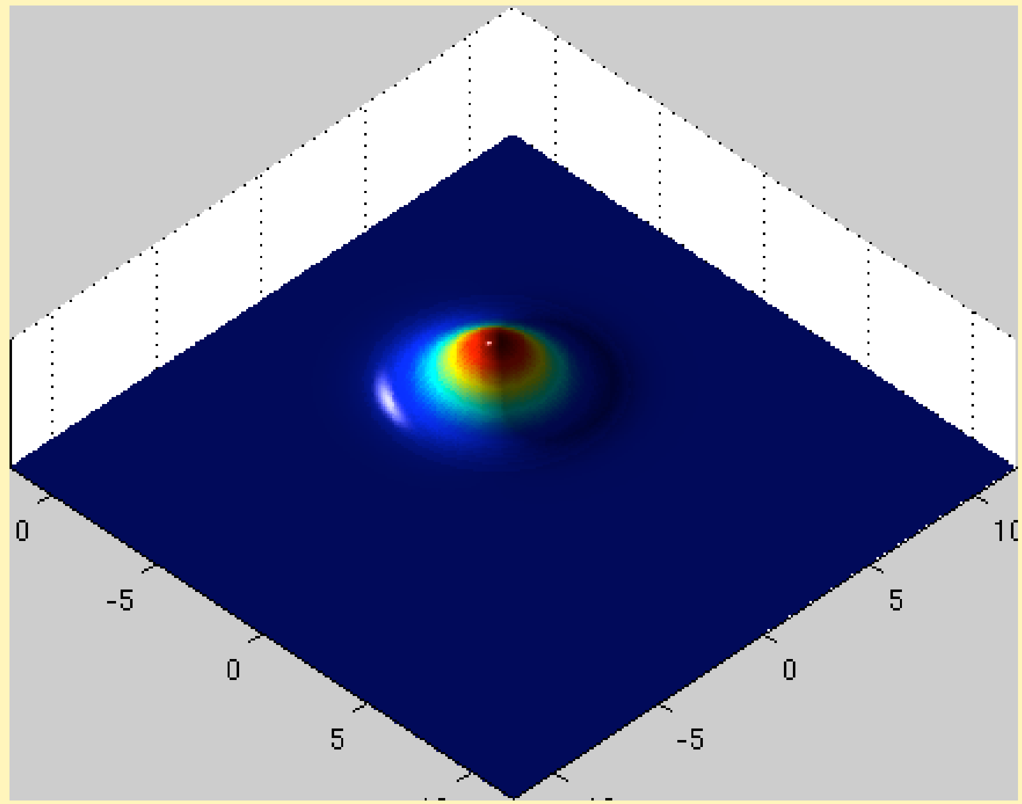
*Other shapes are possible:
M. S. Murillo and E. Timmermans, *Contrib. Plasma Physics* **43**, 333 (2003).

Results: Dynamics of Bound Hydrogen Atom

Simulation example:
- hydrogen
- bound state
- Gaussian wavefunction
- 4^{th}-order RK with adaptive step
- further refinements are needed before full implementation

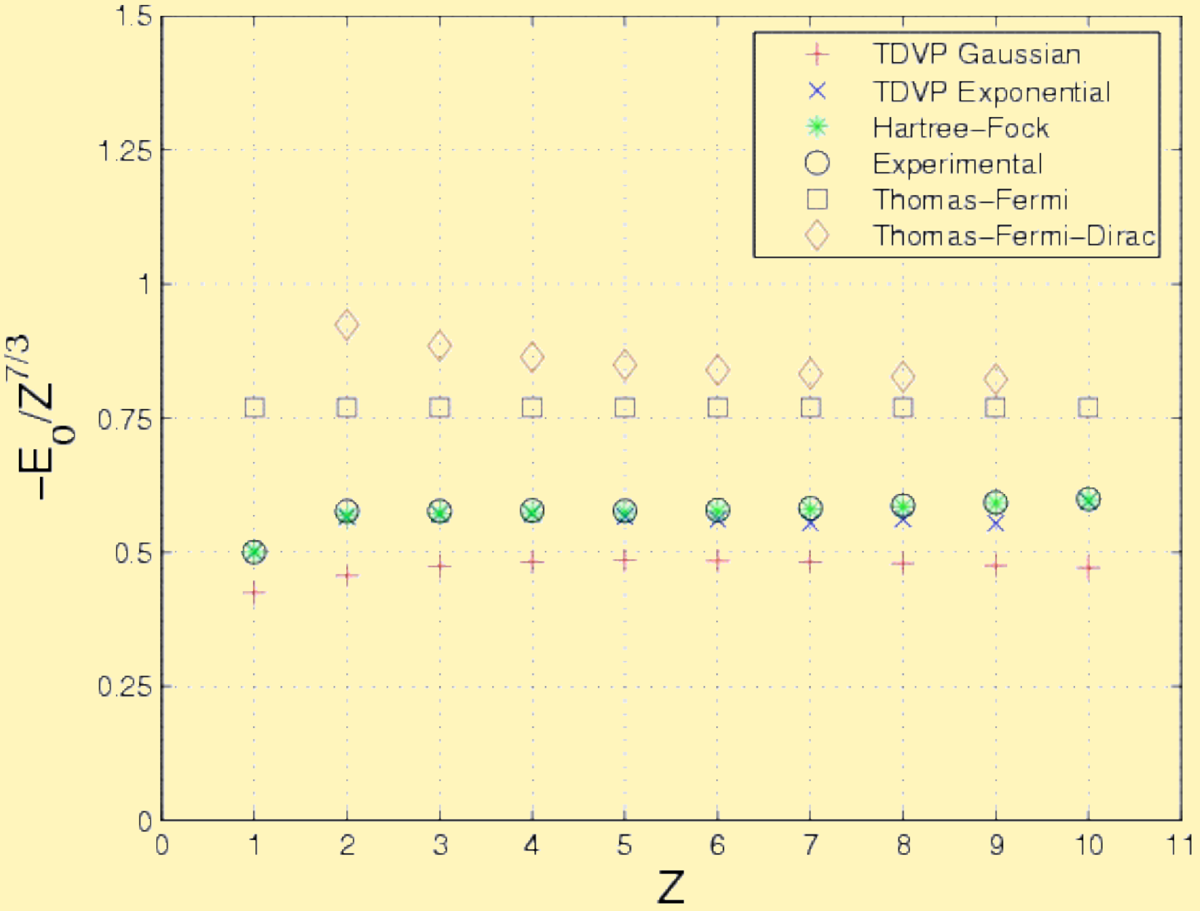


$$|\varphi(x,t)|^2$$



Question: How well can such simple models describe mid-Z projectile atomic physics?

Results: Ground State Energies - Wavepackets Quite Accurate



- Energies compare much more favorably than Thomas-Fermi and Thomas-Fermi-Dirac
- Exponential WP much better than Gaussian, indicating the importance of the cusp at the origin
- These results employ fully antisymmetric total wavefunction – *important physics for plasma degeneracy*

Results: We Have Studied Ground-State Densities

Excellent ground states are found for lower-Z elements, with exponential shapes somewhat better.