Dynamics of fast molecular ions in solids and plasmas

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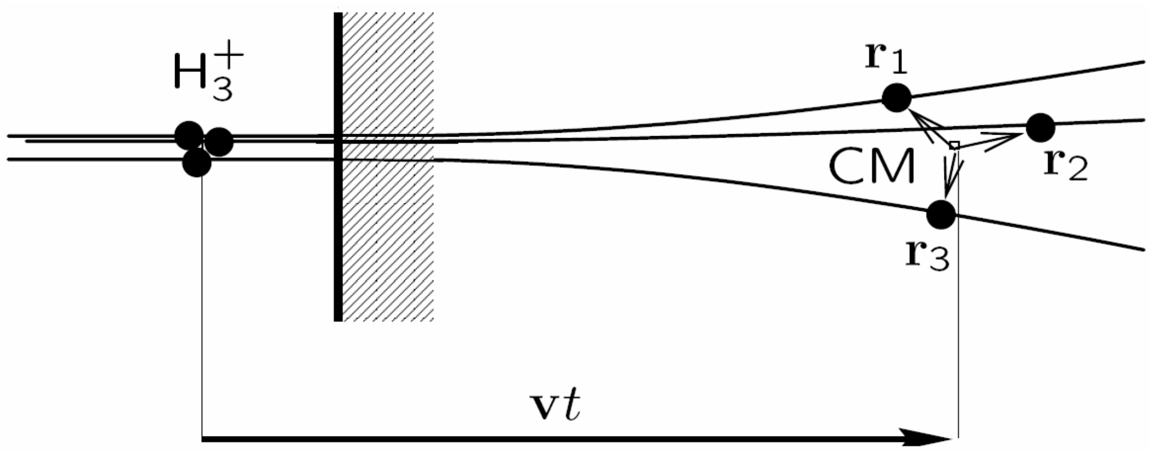
Support: NSERC, PREA, NNSFC, NEMC

Outline

- Introduction
- Charge states
- Coulomb explosions
  - solids
  - plasmas
  - effects of laser
- Grazing scattering from surfaces
- More exotic cases
  - Dust particles in complex plasmas
  - Channeling through carbon nanotubes
- Outlook

Cluster penetration through target



- Fast clusters: $v > v_{Bohr}$ or $v > v_{Therm}$
- Adiabatic evolution of structure in CM frame: $\left|\frac{d\mathbf{r}_j}{dt}\right| << v$
- Rapid ionization (< 1 fs) followed by charge quasi-equilibrium: $\frac{d}{dt}\langle Q_j \rangle \approx 0$
- Vicinage effects due to $\left|\mathbf{r}_j - \mathbf{r}_l\right| \propto v / \omega_{res}$ affect:
  - Cluster stopping power
  - Coulomb explosion
  - Ion charge states

Dielectric theory of cluster stopping power

Review: N.R. Arista, NIMB 164-165 (2000) 108

Decomposition $S_{cl} = S_{incoh} + S_{coh}$ gives stopping ratio $\mathcal{R}_S = 1 + \frac{S_{coh}}{S_{incoh}}$

$$S_{incoh} = \sum_{j=1}^{n} \int \frac{d^3\mathbf{k}}{2\pi^2} |\rho_j(\mathbf{k})|^2 \frac{\mathbf{k}\cdot\mathbf{v}}{k^2\, v} \mathrm{Im}\left[\frac{-1}{\epsilon(k, \mathbf{k}\cdot\mathbf{v})}\right]$$

$$S_{coh} = \sum_{j=1}^{n} \sum_{j\neq l=1}^{n} \int \frac{d^3\mathbf{k}}{2\pi^2} \rho_j(\mathbf{k}) \rho_l^*(\mathbf{k})\, e^{i\mathbf{k}\cdot\mathbf{r}_{jl}} \frac{\mathbf{k}\cdot\mathbf{v}}{k^2\, v} \mathrm{Im}\left[\frac{-1}{\epsilon(k, \mathbf{k}\cdot\mathbf{v})}\right]$$

Charge densities (form factors) of the *j*-th and *l*-th ions; interference factor

Because ion sizes are $<< \left|\mathbf{r}_{jl}\right|$, one can use point-charge approximation in coherent stopping, $\rho_j(\mathbf{k}) \approx Q_j$ and $\rho_l(\mathbf{k}) \approx Q_l$, but **not** in incoherent stopping

Target polarization effects on Coulomb explosion

Newton's equations in CM: $$m_l \frac{d^2\mathbf{r}_l}{d\tau^2} = \mathbf{F}_l^s + \sum_{l \neq j = 1}^{n} \mathbf{F}_{jl}$$

Stopping force on the *l*-th ion

Force on the *l*-th ion from the j-th ion: $$\mathbf{F}_{jl} = -\frac{\partial V_{jl}(\mathbf{r}_{jl})}{\partial \mathbf{r}_l}$$

Point-charge approximation in the inter-ion potential: $\rho_j(\mathbf{k}) \approx Q_j$ and $\rho_l(\mathbf{k}) \approx Q_l$

$$V_{jl}(\mathbf{r}_{jl}) = \int \frac{d^3\mathbf{k}}{(2\pi)^3} \frac{4\pi}{k^2} \frac{\rho_j(\mathbf{k})\rho_l^*(\mathbf{k})}{\epsilon(k, \mathbf{k}\cdot\mathbf{v})} \mathrm{e}^{i\mathbf{k}\cdot(\mathbf{r}_l - \mathbf{r}_j)}$$

Charge on isolated "heavy" ion: Brandt-Kitagawa model

solids: W. Brandt and M. Kitagawa, PRB 25 (1982) 5631

plasmas: J. D'Avanzo *et al*., Phys. Plasmas 3 (1996) 3885

$$E_{iso} = c_k \int d^3\mathbf{r}\,\rho^{5/3}(\mathbf{r}) - \int d^3\mathbf{r}\,\rho(\mathbf{r})\frac{Z}{r} + \frac{\lambda}{2}\iint d^3\mathbf{r}\,d^3\mathbf{r}'\,\frac{\rho(\mathbf{r})\rho(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}$$

Energy of *N* bound elecrons in LAB frame: $E_L = N\frac{v_r^2}{2} + E_{iso}(N)$

Stripping criterion: $\frac{dE_L}{dN} \equiv \frac{v_r^2}{2} + E'_{iso}(N_0) = 0$

Charge on isolated ion in dynamic quasi-equilibrium:

$$Q_0(v) \equiv Z - N_0$$

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- Outlook

Vicinage effect on ion charges in cluster with n ions

Experiment:

- A. Brunelle, S. Della-Negra, J. Depauw, D. Jacquet, Y. Le Beyec, and M. Pautrat, *Phys. Rev. A*, **59** (1999) 4456.
- A. Chiba, Y. Saitoh, and S. Tajima, *Nucl. Instrum. Methods Phys. Res. B*, **232** (2005) 32.

Theory:

1] Z. L. Mišković, S. G. Davison, F. O. Goodman, W.-K. Liu, and Y.-N. Wang, *Phys. Rev. A*, **61** (2000) 62901.

2] J.W. Hartman, T.A. Tombrello, S. Bouneau, S. Della Negra, D. Jacquet, Y. Le Beyec, and M. Pautrat, *Phys. Rev. A*, **62** (2000) 43202.

3] S. Heredia-Avalos, R. Garcia-Molina, and N.R. Arista, *Europhys. Lett.*, **54** (2001) 729.

4] E.Nardi, Z. Zinamon, T.A. Tombrello, and N.M Tanushev, *Phys. Rev. A*, **66** (2002) 13201.

5] T. Kaneko, *Phys. Rev. A*, **66** (2002) 52901.

Vicinage effect on ion charges in cluster with n ions

Z.L. Miskovic *et al*., PRA 63 (2001) 22901

Total electronic energy in cluster of n ions in LAB frame:

$$\mathcal{E}_{\mathcal{L}} = \sum_{j=1}^{n} N_j \frac{v_r^2}{2} + \sum_{j=1}^{n} E_{iso}(N_j) + \frac{1}{2} \sum_{j=1}^{n} \sum_{l \neq j}^{n} (Z_j - N_j)(Z_l - N_l) U\left(\mathbf{r}_{jl}\right)$$

Point-charge approximation in the inter-ion potential: $\rho_j(\mathbf{k}) \approx Q_j = Z_j - N_j$

$$U(\mathbf{r}) = \int \frac{d^3\mathbf{k}}{(2\pi)^3} \frac{4\pi}{k^2} \frac{e^{i\mathbf{k}\cdot\mathbf{r}}}{\epsilon(k, \mathbf{k}\cdot\mathbf{v})}$$

(We use both Yukawa potential with v/ω_{res}, and potential exhibiting wake)

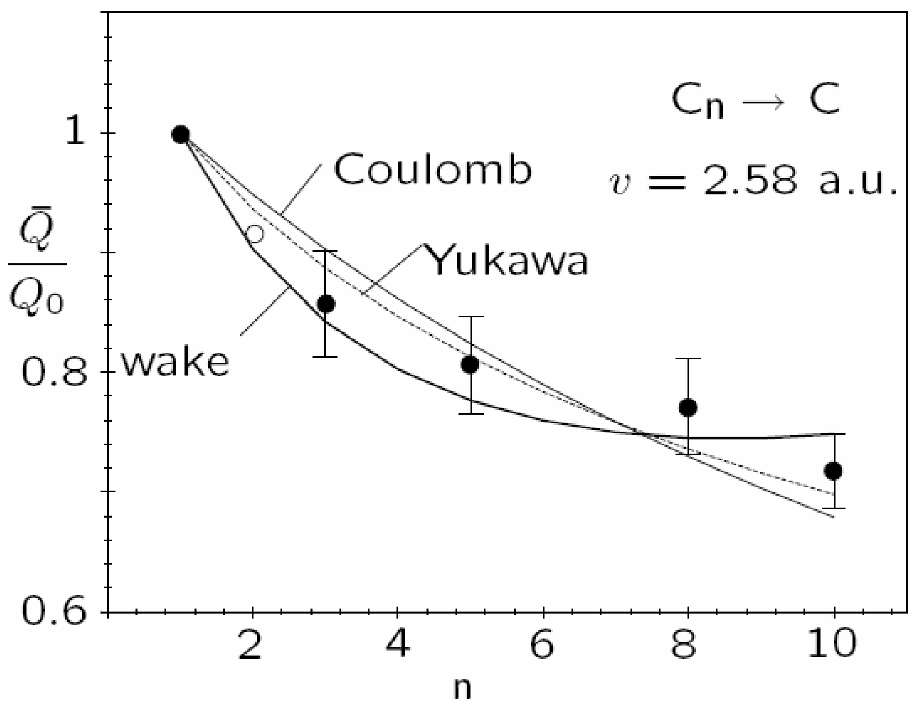
Stripping criterion $\frac{\partial \mathcal{E}_{\mathcal{L}}}{\partial N_j} = 0$ gives n equations for $N_1, N_2, \ldots, N_n$:

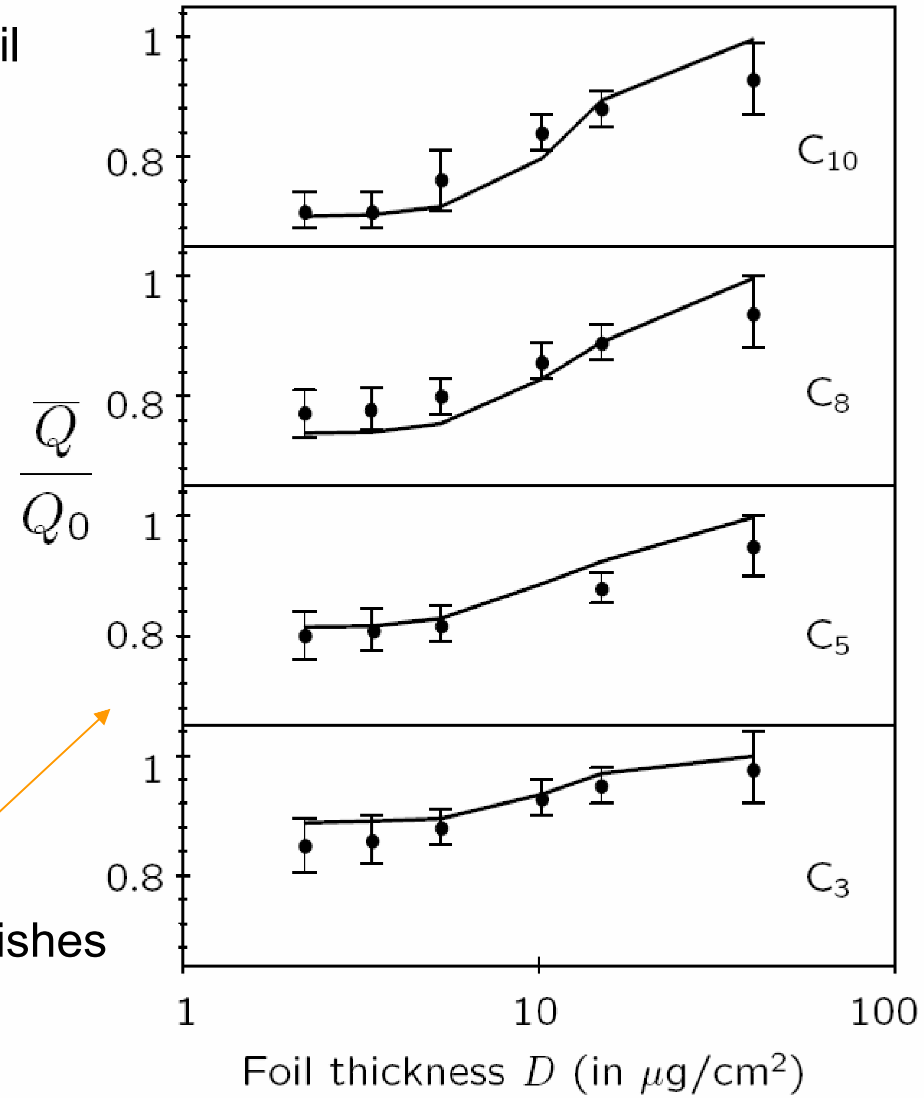
$$\frac{v_r^2}{2} + E'_{iso}(N_j) - \sum_{j \neq l = 1}^{n} (Z_l - N_l)\, U\left(\mathbf{r}_{jl}\right) = 0$$

Vicinage effect on ion charges for $C_n^+ \rightarrow$ carbon foil

A. Brunelle *et al.*, PRA 59 (1999) 4456; & A. Chiba *et al.*, NIMB 232 (2005) 32

Charge reduction vs cluster size in thin foil



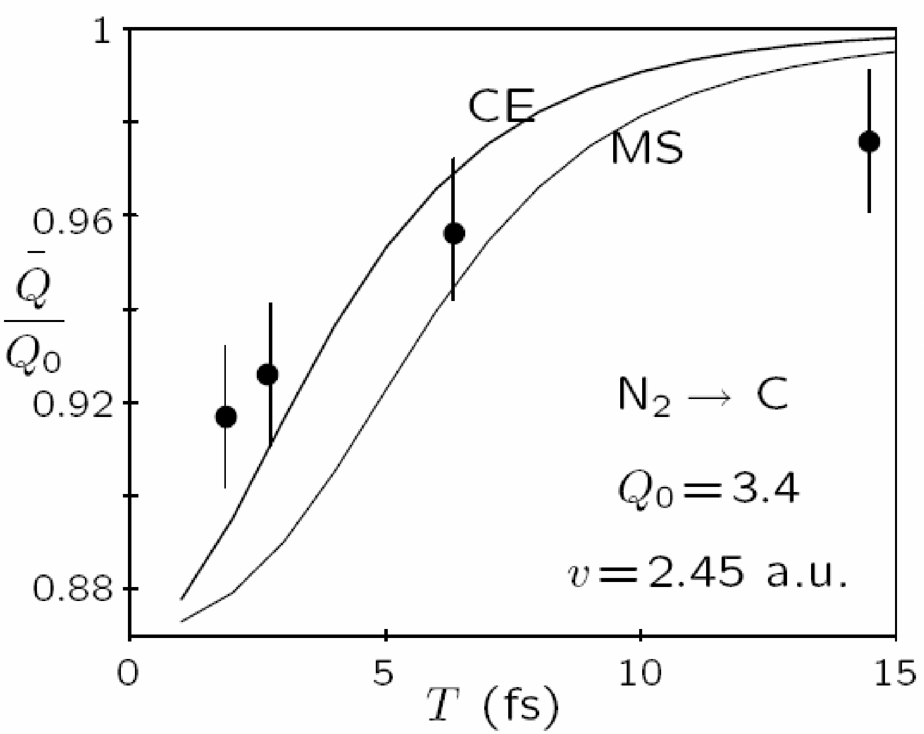


Vicinage effect on ion charges diminishes with increasing foil thickness due to **multiple scattering** on target atoms

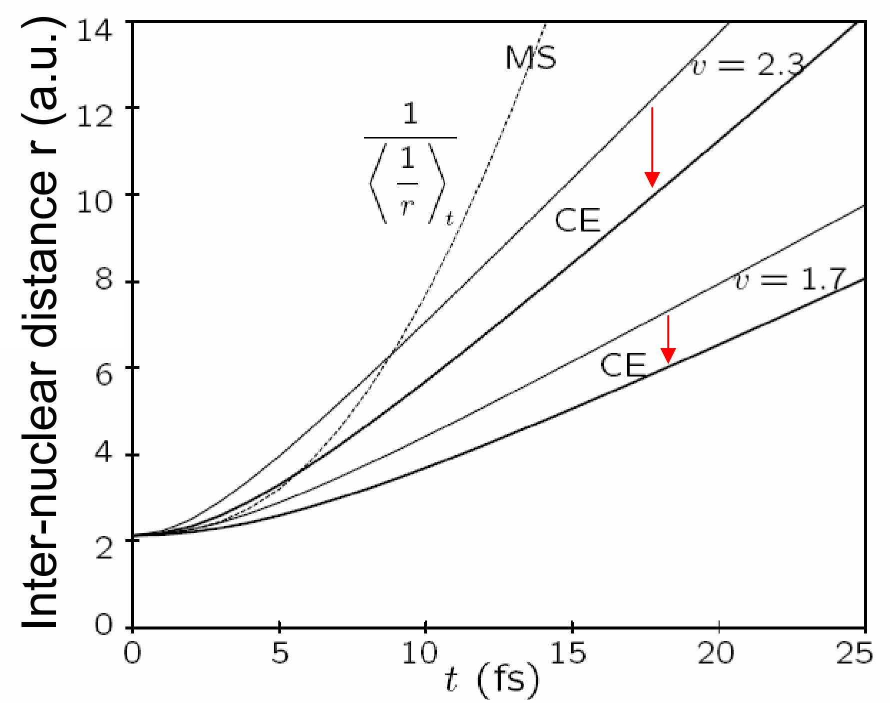
Vicinage effect on ion charges for $N_2^+ \rightarrow$ carbon foil

D. Maor *et al*., PRA 32 (1985) 105

Charge reduction vs dwell time due to Coulomb explosion (CE) **or** multiple scattering (MS)



Charge reduction slows down Coulomb explosion (CE), but **not** multiple scattering (MS)

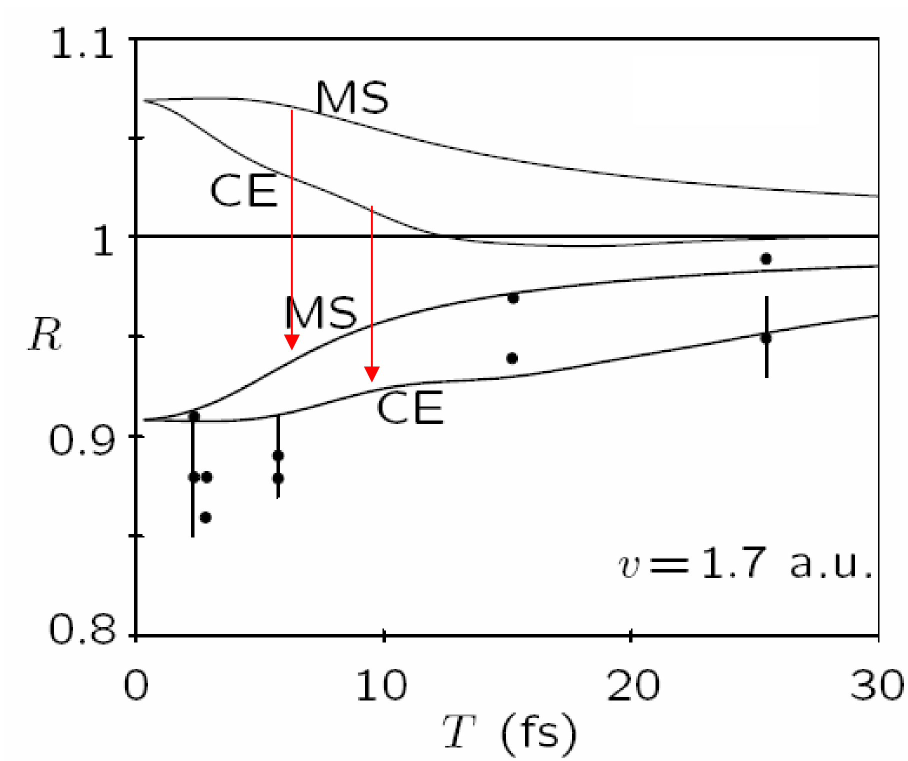


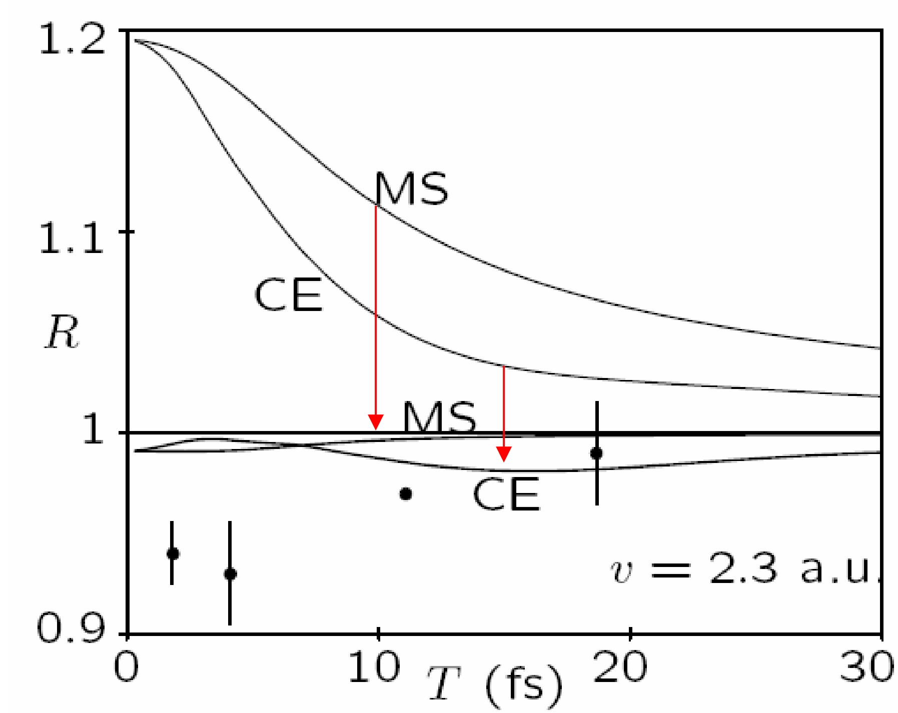
(Charge states and Coulomb explosions evaluated with Yukawa-type potential)

Vicinage effect on ion charges affects energy losses for fast N_2^+ in carbon foil

M.F. Steuer *et al.*, IEEE Trans. Nucl. Sci., NS-30 (1983) 1069

Conclusion: charge reduction can change stopping power ratio from *R* > 1 to *R* < 1





Alternative explanation: R. Garcia-Molina & S. Heredia-Avalos, PRA 63 (2001) 44901

Ion charge distribution in **large** homo-nuclear clusters made of heavy ions, passing through very thin foils

Total electronic energy in cluster of n ions in LAB frame:

$$\langle \mathcal{E}_{\mathcal{L}} \rangle [q(\mathbf{r})] = \frac{v_r^2}{2} \int d^3\mathbf{r} \left(Z - q(\mathbf{r}) \right) f_1(\mathbf{r}) + \int d^3\mathbf{r}\, E_{iso}[q(\mathbf{r})]\, f_1(\mathbf{r}) + \frac{1}{2} \iint d^3\mathbf{r}\, d^3\mathbf{r}'\, q(\mathbf{r}) q(\mathbf{r}')\, U(\mathbf{r} - \mathbf{r}') f_2(\mathbf{r}, \mathbf{r}')$$

Statistical description of cluster structure: $f_2(\mathbf{r}, \mathbf{r}') = f_1(\mathbf{r}) f_1(\mathbf{r}') g_2(|\mathbf{r} - \mathbf{r}'|)$

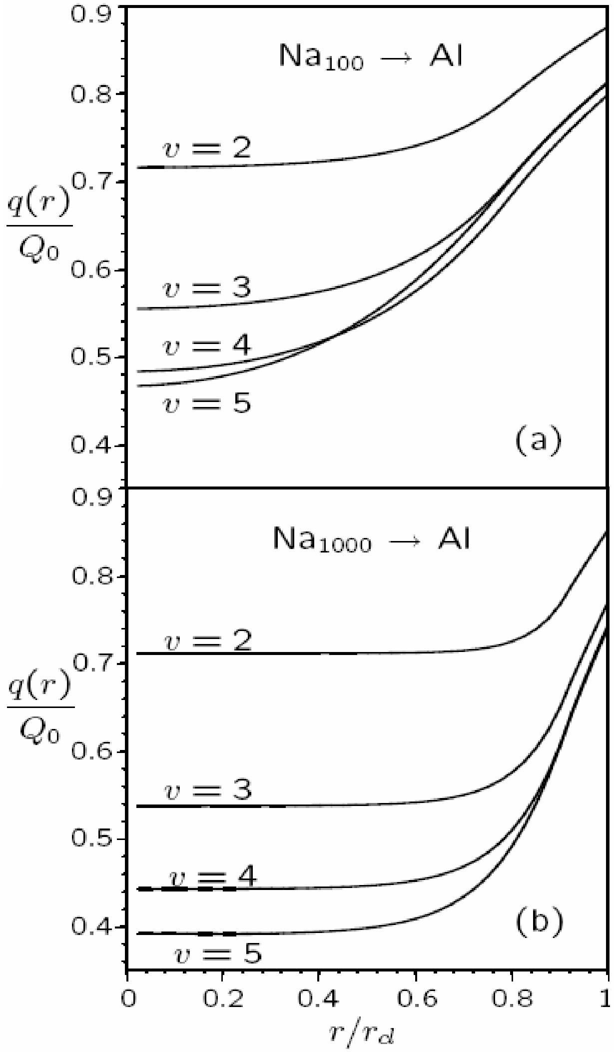
Stripping criterion $\dfrac{\delta \langle \mathcal{E}_{\mathcal{L}} \rangle}{\delta q(\mathbf{r})} = 0$ gives integral equation for charge distribution $q(\mathbf{r})$:

$$-\frac{v_r^2}{2} + E'_{iso}(q(\mathbf{r})) + (\mathrm{n} - 1) \int d^3\mathbf{r}'\, q(\mathbf{r}')\, f_1(\mathbf{r}')\, g_2(|\mathbf{r} - \mathbf{r}'|)\, U(\mathbf{r} - \mathbf{r}') = 0$$

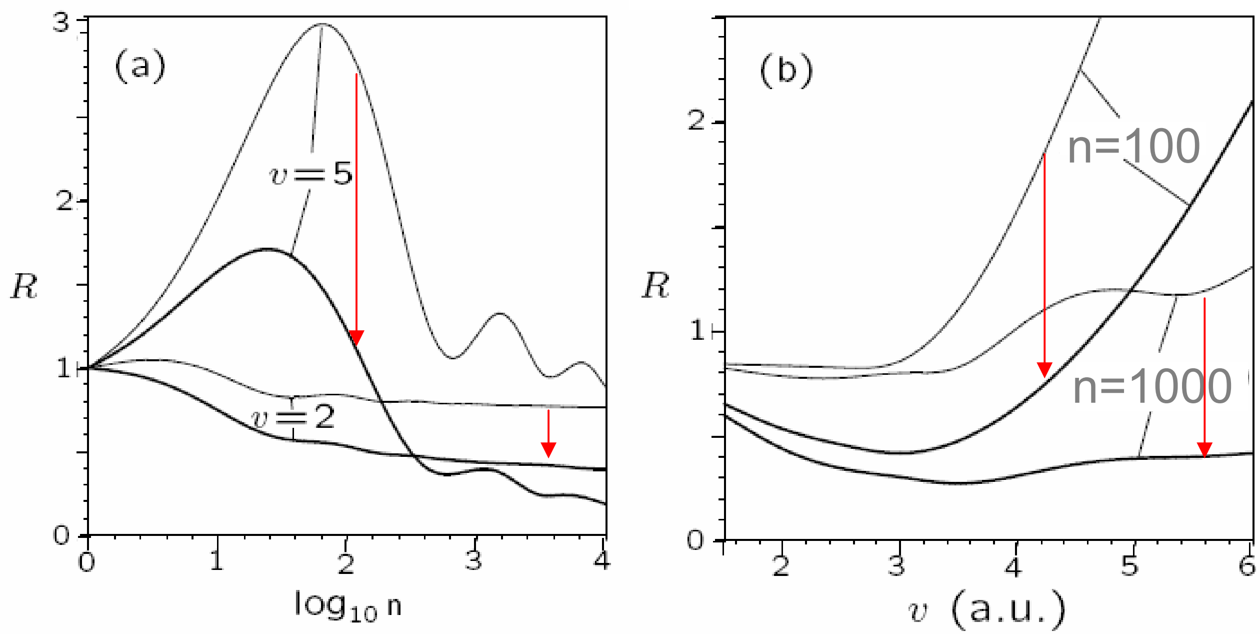
Ion charges and stopping ratio for $Na_n^+ \rightarrow Al$

Z.L. Miskovic *et al.*, PRA 67 (2003) 22903

Charge distribution through cluster



Conclusion: non-homogeneous charge distribution *q(r)* can reduce stopping power ratio *R* compared to the case of homogeneous distribution where $q(r) = Q_0 = const.$ is assumed



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- ❑ **Introduction**
- ❑ **Charge states**
- ❑ **Coulomb explosions**
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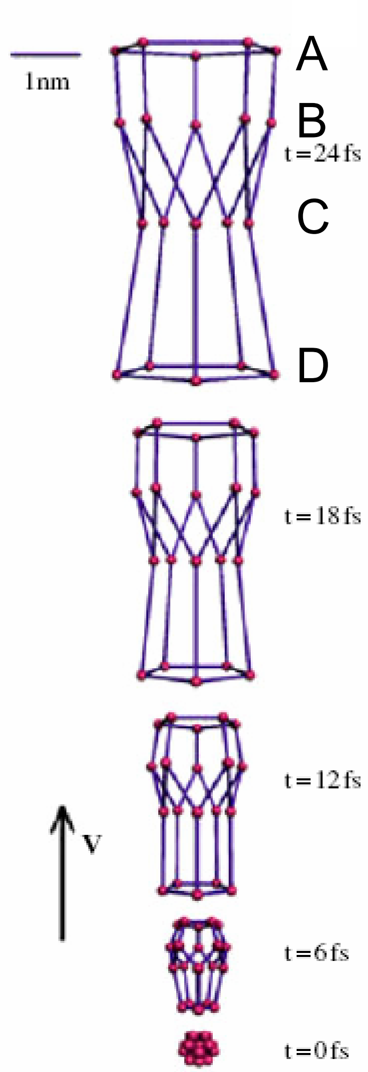
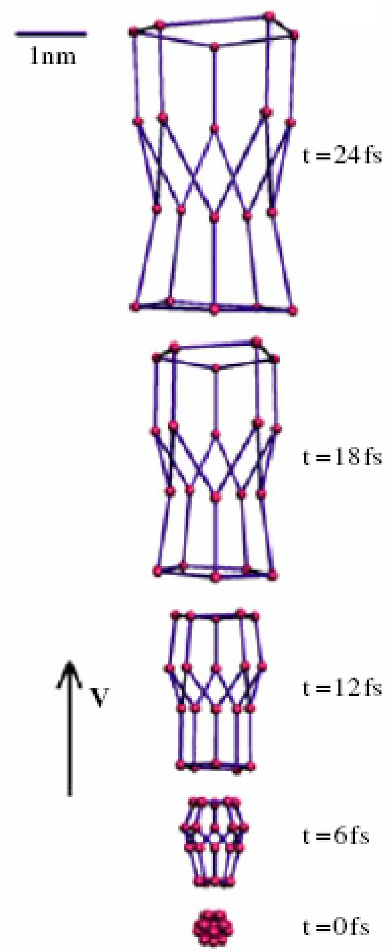
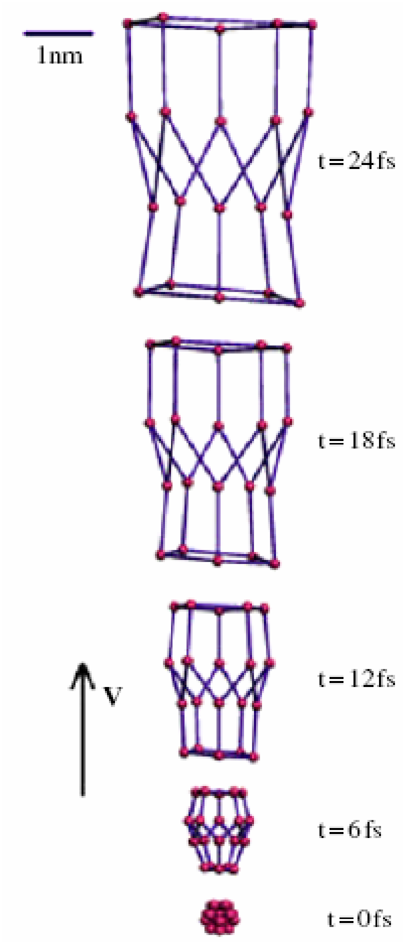
Charge states, Coulomb explosion, multiple scattering and energy loss of C_{20}^{+} in Al foil

H.W. Li *et al*., J. Phys.: Cond. Matt. 16 (2004) 1231





Multiple scattering
included by a MC sim.



All ion charges
frozen at Q_{iso}

Charge states, Coulomb explosion, multiple scattering and energy loss of C_{20}^{+} in Al foil

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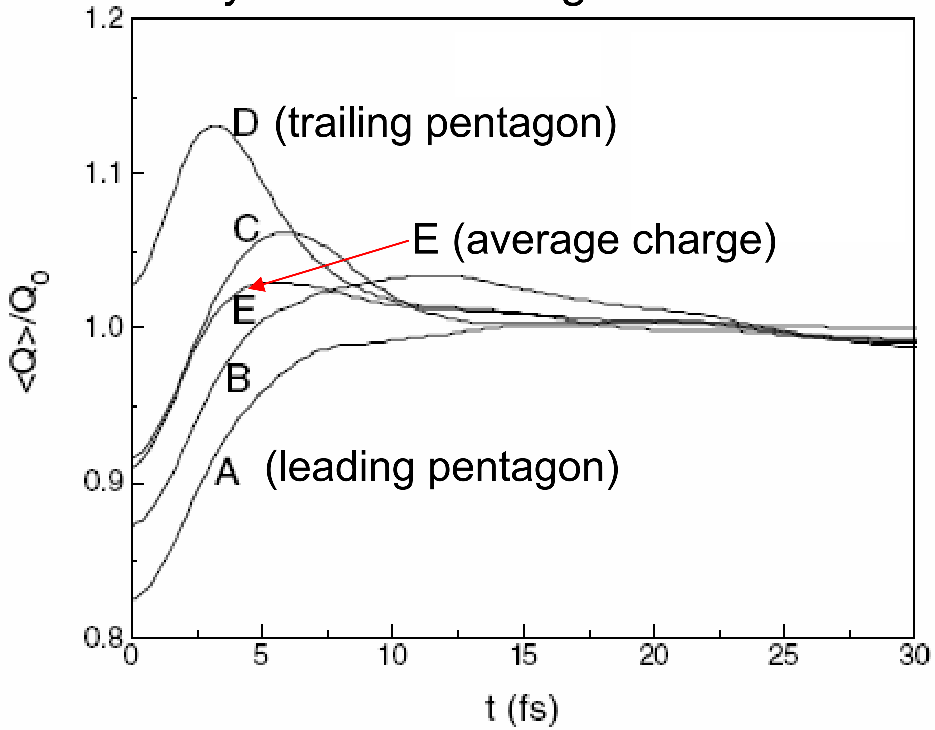
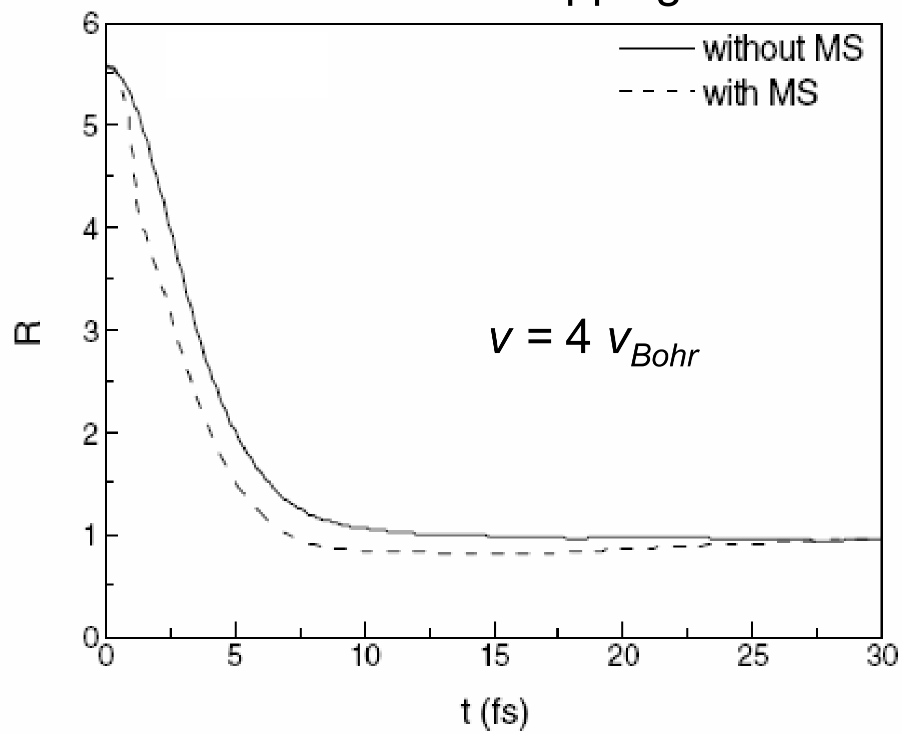
Wake in inter-ionic potential gives asymmetrical charge distribution



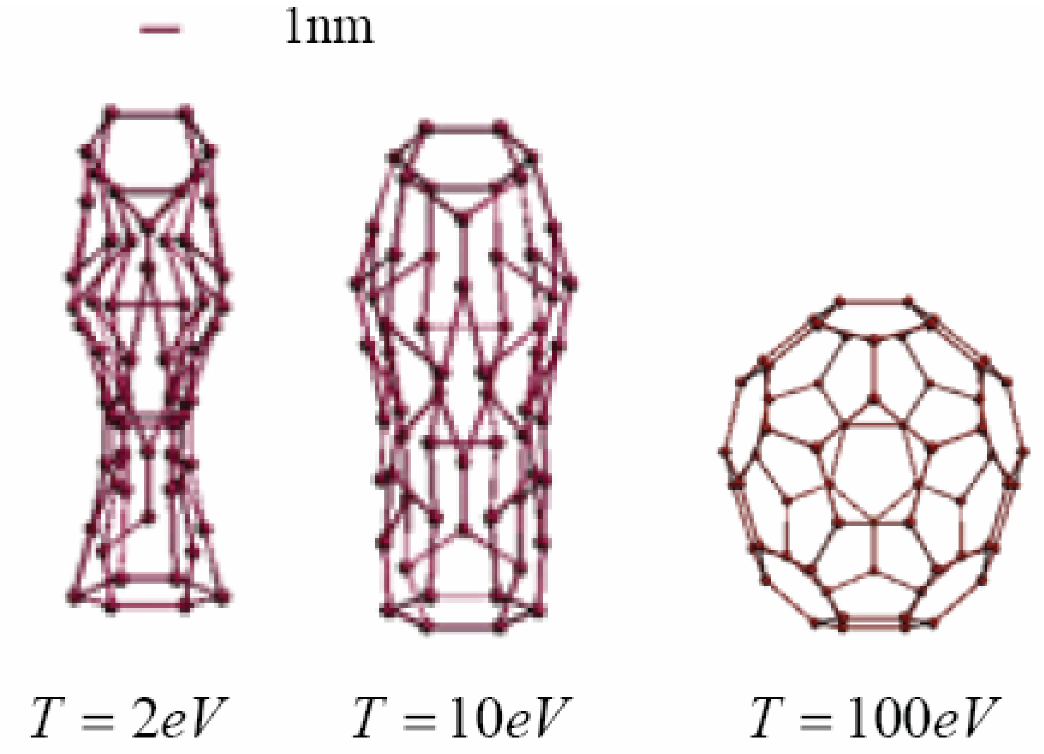
Effects of multiple scattering on reduction of stopping ratio



Coulomb explosion of C_{60}^{+} in hot dense plasma

G.Q. Wang *et al*., Phys. of Plasmas 12 (2005) 42702

Effects of electron temperature after 25 fs

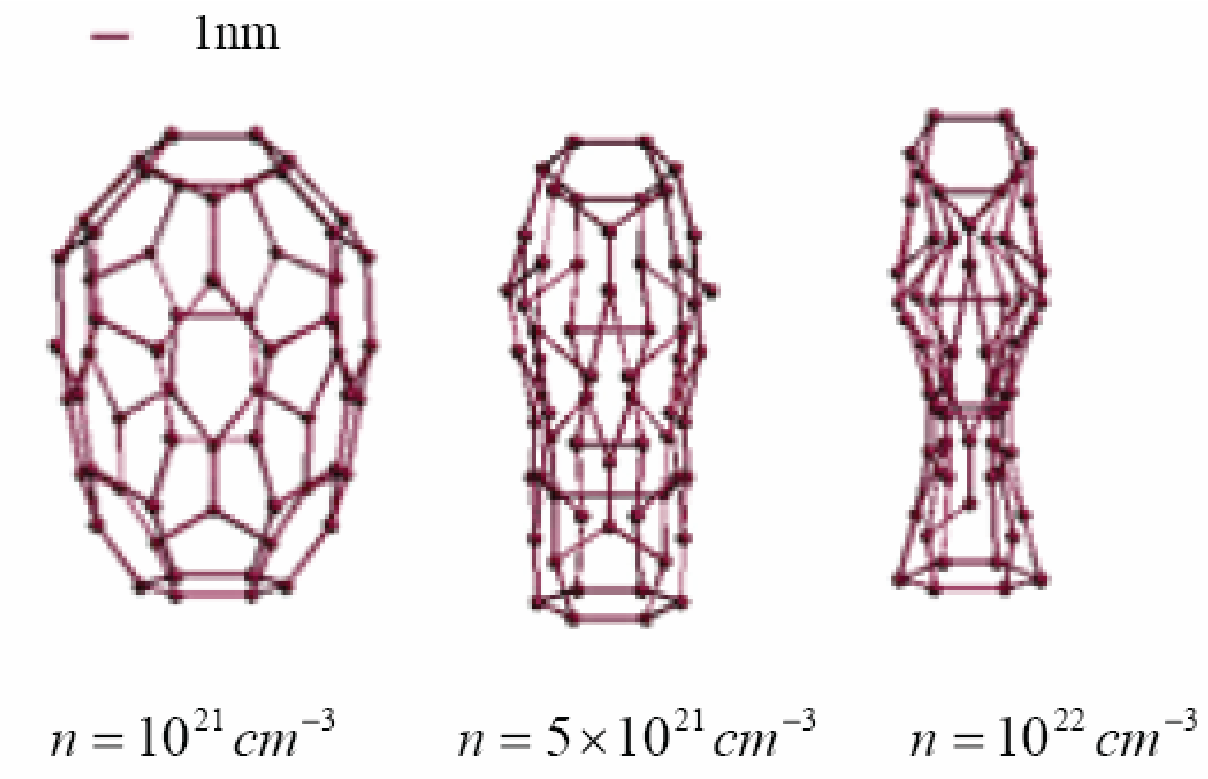


Other parameters: plasma density $n = 10^{22}$ cm^{-3}, cluster speed $v = 3\ v_T$

Coulomb explosion of C_{60}^{+} in hot dense plasma

G.Q. Wang *et al*., Phys. of Plasmas 12 (2005) 42702

Effects of <u>plasma density</u> after 25 fs

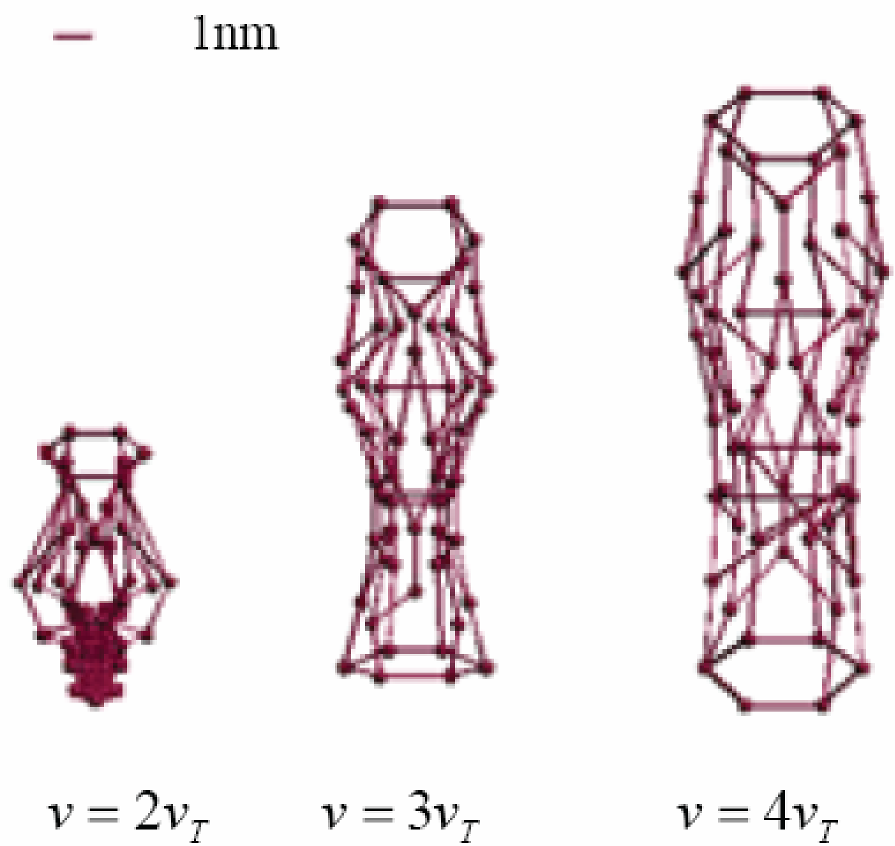


Other parameters: electron temperature T = 2 eV, cluster speed $v = 3\ v_T$

Coulomb explosion of C_{60}^{+} in hot dense plasma

G.Q. Wang *et al*., Phys. of Plasmas 12 (2005) 42702

Effects of cluster speed after 25 fs



Other parameters: plasma density $n = 10^{22}$ cm^{-3}, electron temperature T = 2 eV

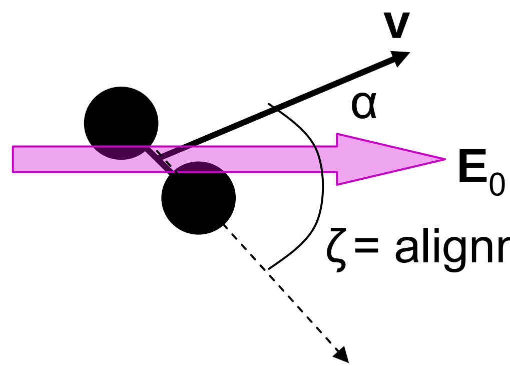
Coulomb explosion of H_2^+ in laser-ablated Al plasma

experiments: A. Sakumi et al, NIMA 415 (1998) 648

theory: G.Q. Wang *et al*., PRA 66 (2002) 42901

Effects of laser on molecule **alignment**

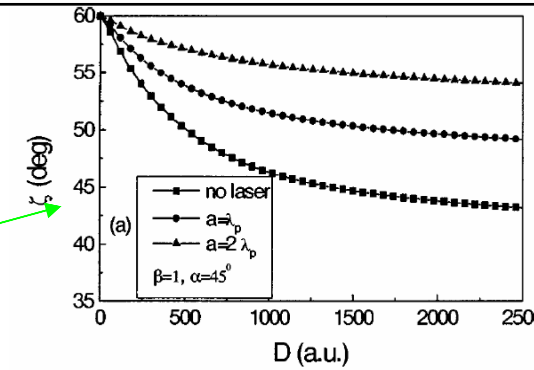
Effects:
- laser intensity $a = E_0 / \omega_0^2$
- laser frequency $\beta = \omega_0 / \omega_p$
- laser angle α

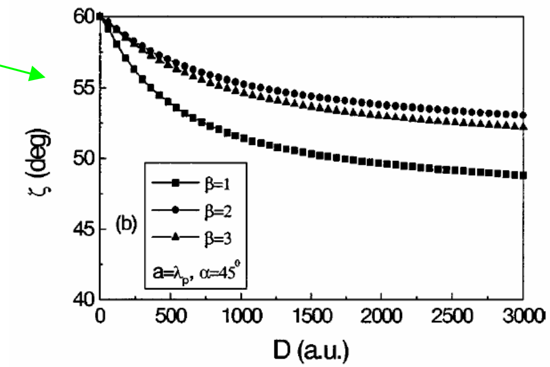


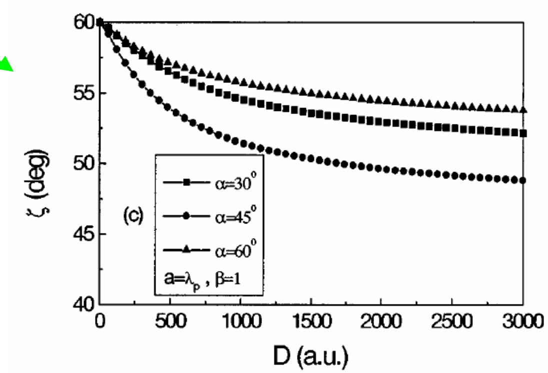
ζ = alignment angle

v = 3 a.u.

ζ_0 = 60°





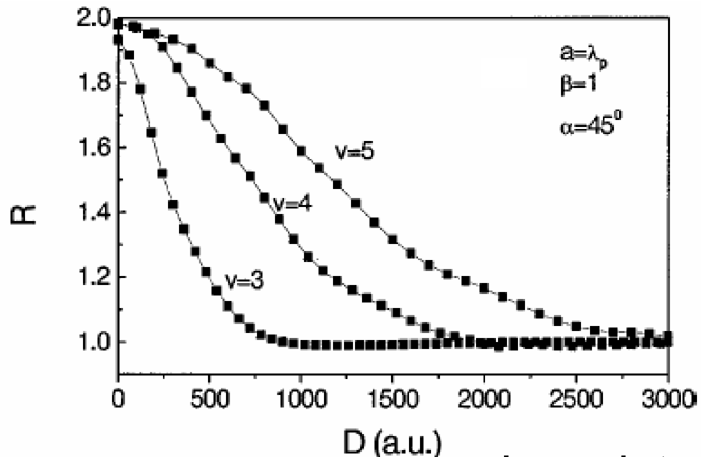


Coulomb explosion of H_2^+ in laser-ablated Al plasma

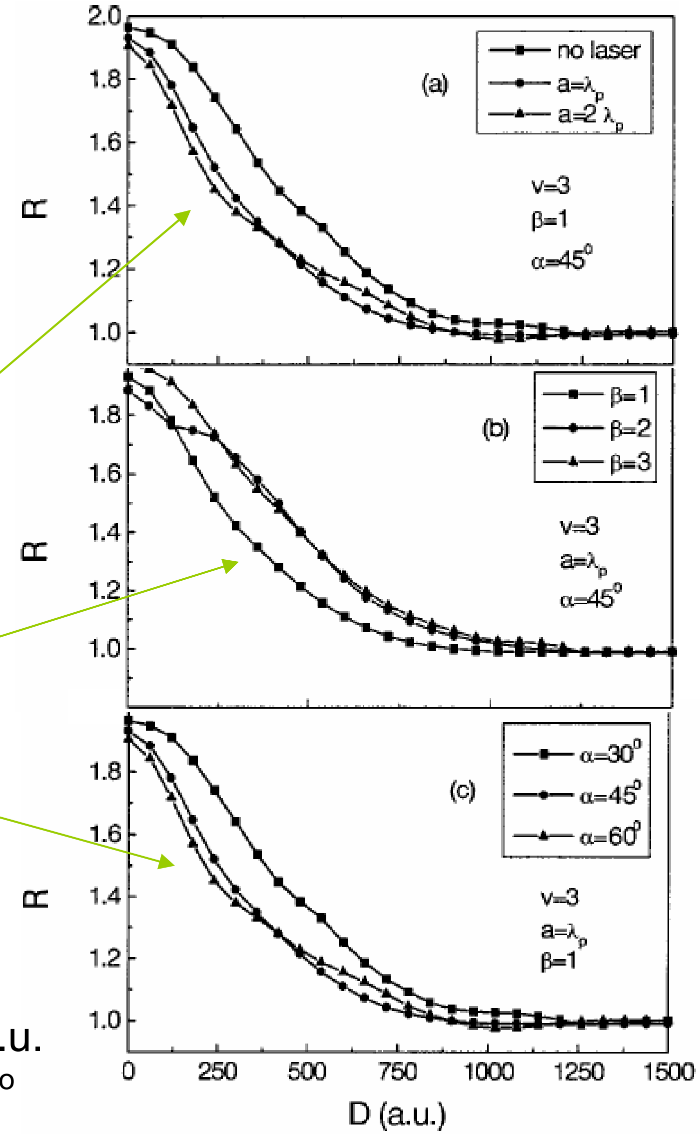
experiments: A. Sakumi *et al*, NIMA 415 (1998) 648

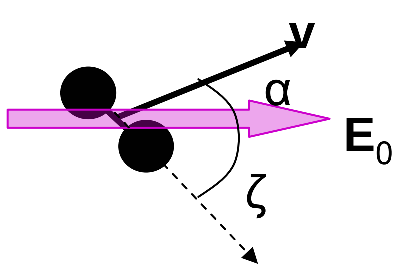
theory: G.Q. Wang *et al*., PRA 66 (2002) 42901

Effects of cluster speed (in a.u.) on **stopping ratio** vs penetration depth



Effects:
- laser intensity $a = E_0 / \omega_0^2$
- laser frequency $\beta = \omega_0 / \omega_p$
- laser angle α





v = 3 a.u.

ζ_0 = 60°

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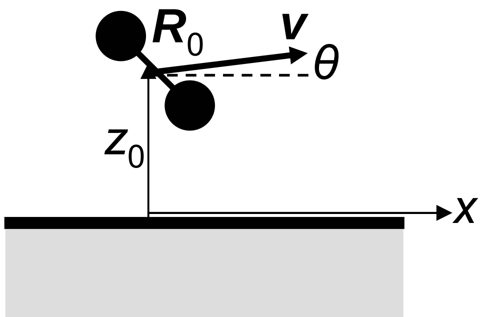
Grazing scattering of N_2^+ from carbon surface

Y.H. Song *et al.*, PRA 72 (2005) 12903



Trajectory of CM: $$\frac{dz_0}{dx_0} = \mp\theta\sqrt{1 - 2\frac{U_{at}(z_0) + U_s(z_0) + U_w(z_0, \mathbf{R}_0)}{E\theta^2}}$$

Relative motion: $$\frac{d\mathbf{R}_0}{dx_0} = \frac{\mathbf{u}}{v} \;, \quad \frac{d\mathbf{u}}{dx_0} = \frac{1}{vm}\left(\mathbf{F}^{(c)} + \mathbf{F}^{(w)}\right)$$

Molecule stopping power: $$S_{mol}(z_0, R_0) = 2S_e(z_0) + S_v(z_0, R_0)$$

Total energy loss $\triangle E_{mol} = \int S_{mol}(z_0, R_0)\, ds$ gives stopping ratio $\Gamma = \frac{\triangle E_{mol}}{2\triangle E_{single}}$

Transition amplitude with step-potential wavefunctions and screened potential *Φ*

$$\mathbf{M}_{\boldsymbol{\kappa}\leftarrow\mathbf{l}_0} = \frac{-i}{(2\pi)^2}\frac{\kappa_z}{\kappa_z + l_z}\left[2\left\langle\varphi^+_{l_z}\left|\phi(\mathbf{Q},\omega,z)\right|\varphi^0_{l^0_z}\right\rangle + \frac{\kappa_z - l_z}{\sqrt{\kappa_z l_z}}\left\langle\varphi^-_{l_z}\left|\phi(\mathbf{Q},\omega,z)\right|\varphi^0_{l^0_z}\right\rangle\right]$$

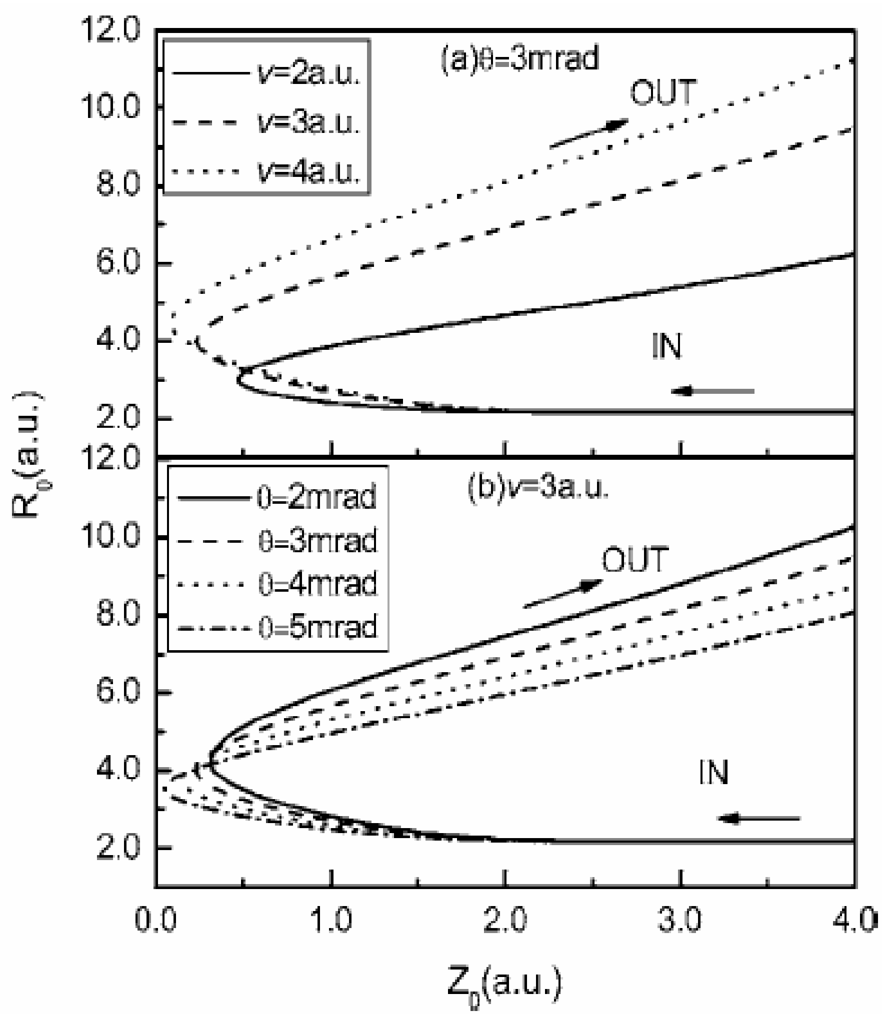
Total electron emission probability: $$W_{mol} = \int \frac{ds}{v}\frac{d^3P}{d\boldsymbol{\kappa}^3}(x_0) = 2W_s + W_v$$

Charge state evolution: $$N(s) = N_\infty - (N_\infty - N_0)\exp\left[-n\sigma_c(s - s_0)\right]$$

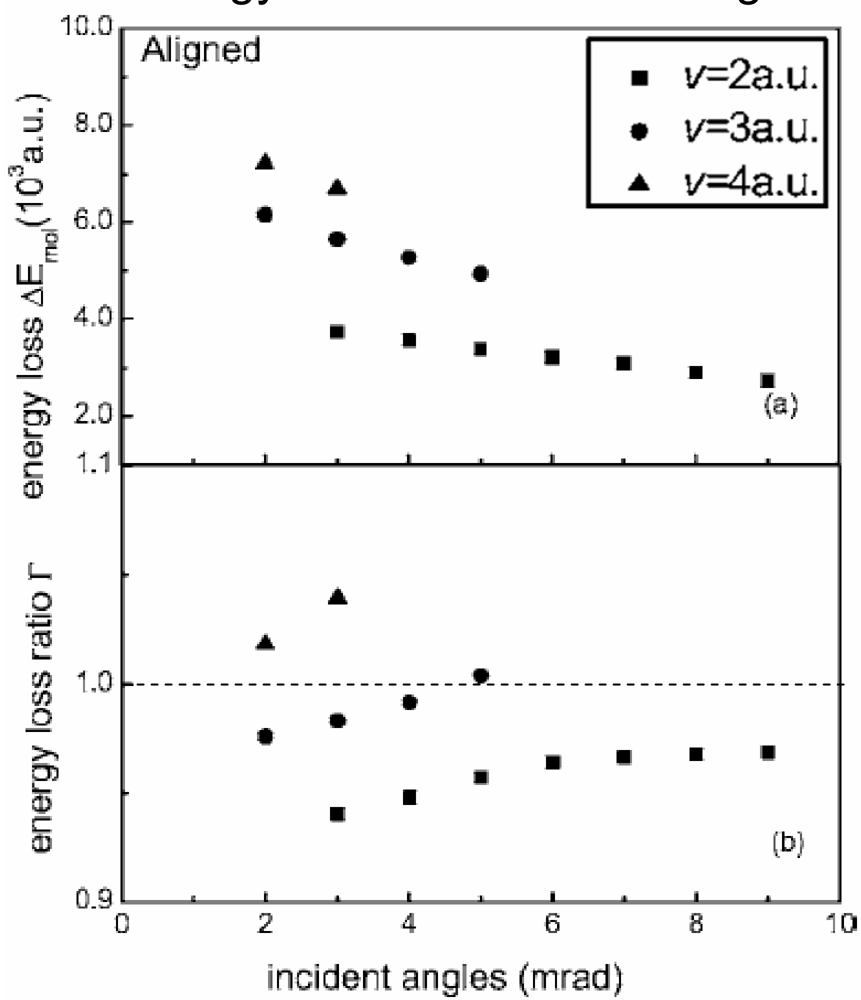
Grazing scattering of N_2^+ from carbon surface

Y.H. Song *et al.*, PRA 72 (2005) 12903

Coulomb explosion of aligned molecules



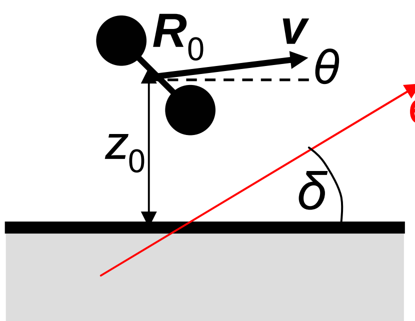
Energy losses vs incident angles



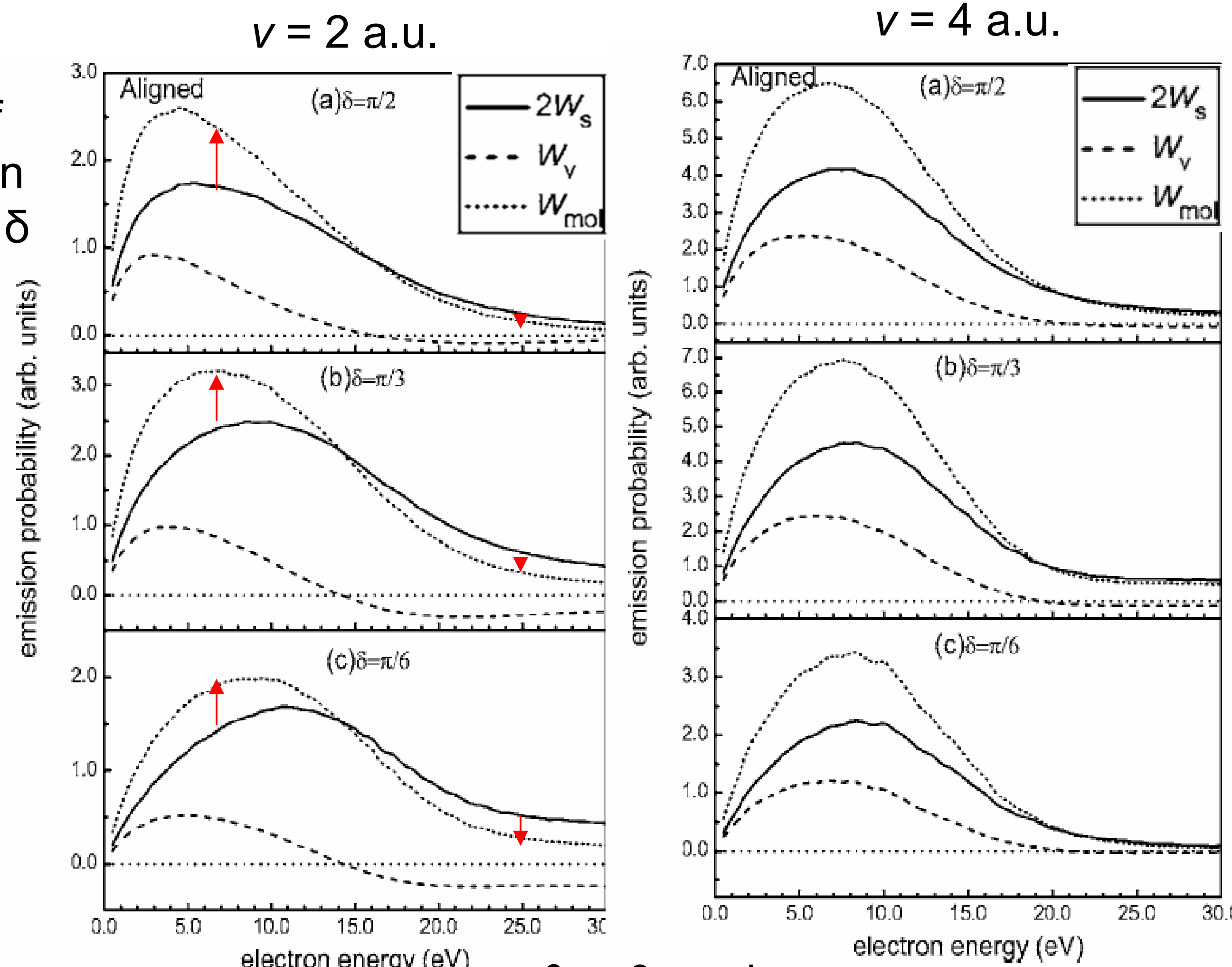
Grazing scattering of N_2^+ from carbon surface

Y.H. Song *et al*., PRA 72 (2005) 12903

Vicinage effects on differential probability of Kinetic electron emission in the direction of angle δ



$$W_{mol} = 2W_s + W_v$$



θ_{in} = 3 mrad

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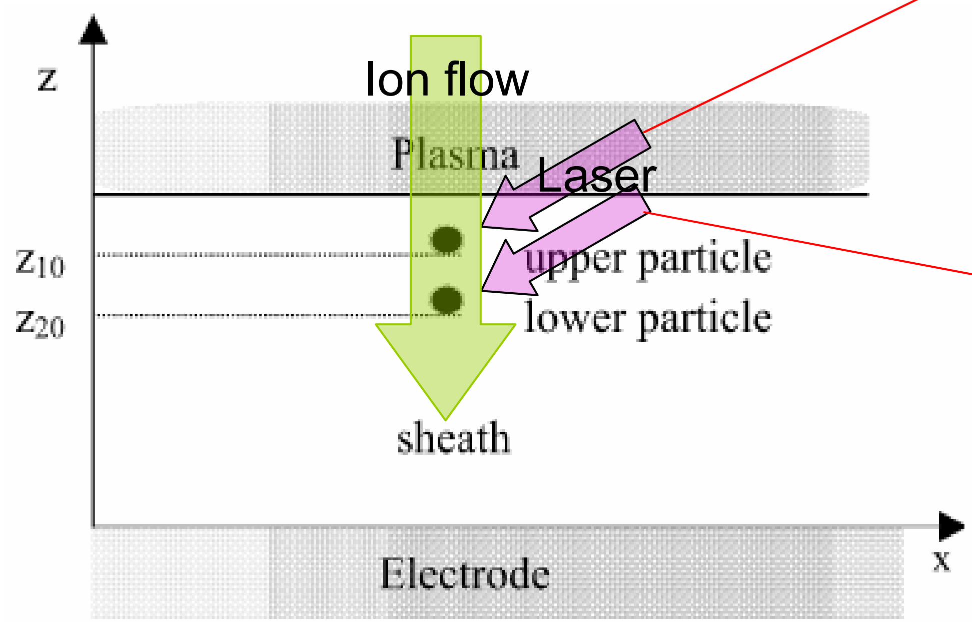
Wake riding effect for dust particles in RF plasma sheath

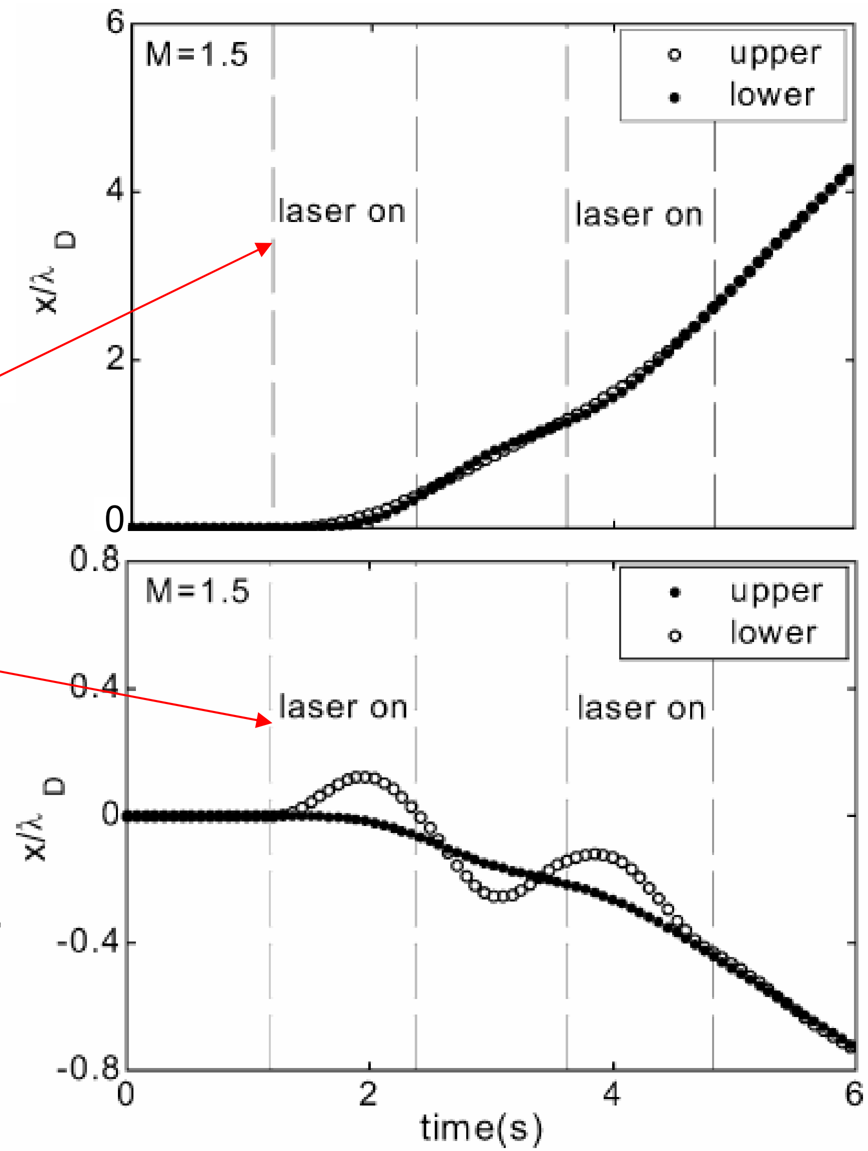
experiment: A.A. Samarian *et al*., Phys. Plasmas, 12 (2005) 22103
theory: L.J. Hou *et al*., PLA 292 (2001) 129

Find horizontal positions of particles from

$$m_i \frac{d^2 x_i}{dt^2} = F_x(x_{ij}, z_{ij}) + F_n(u_{ix}) + F_{\text{opt}}$$

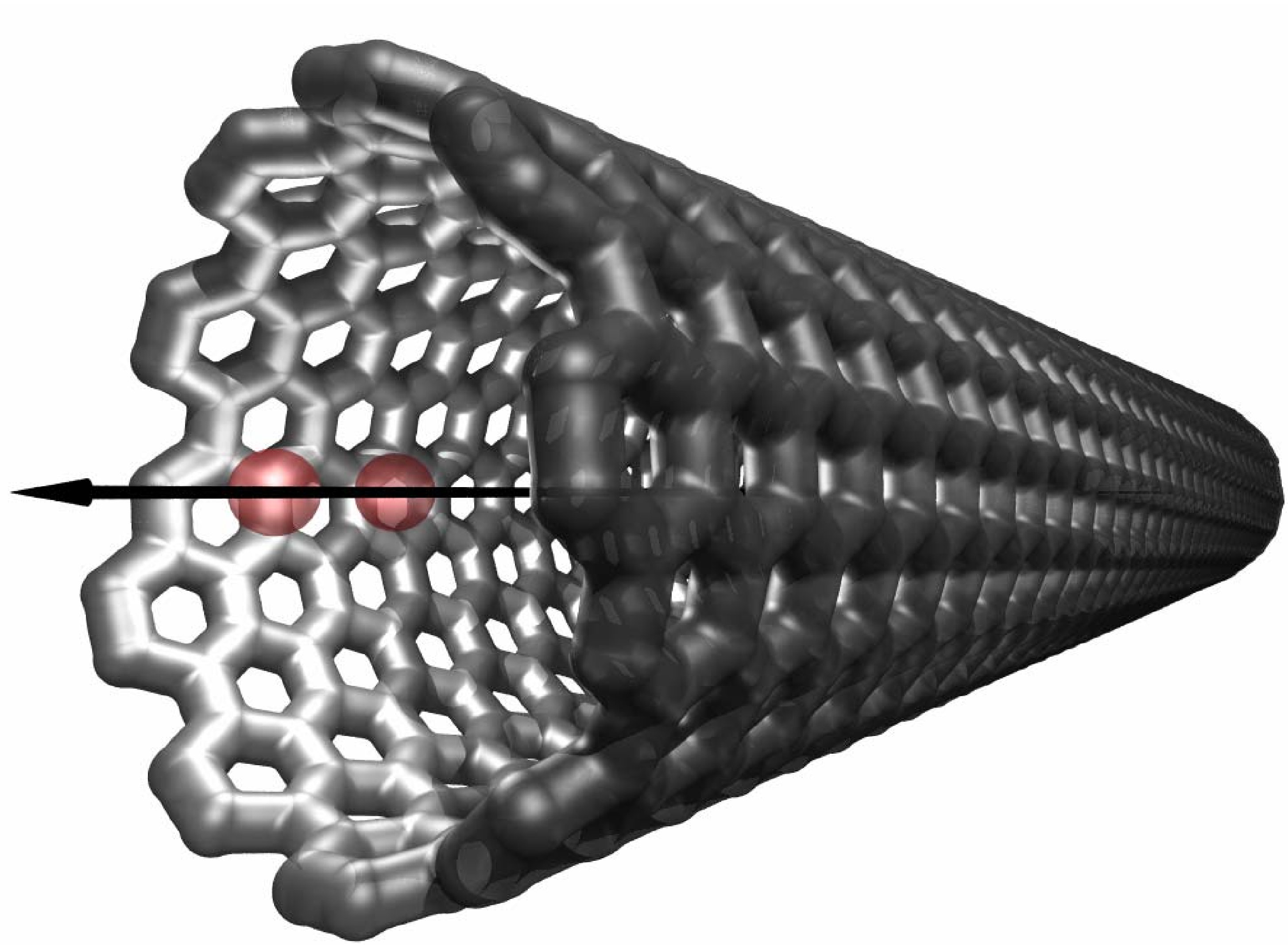
for $z_{ij} = const.$, F_x exhibits asymmetry of wake between upper and lower particles





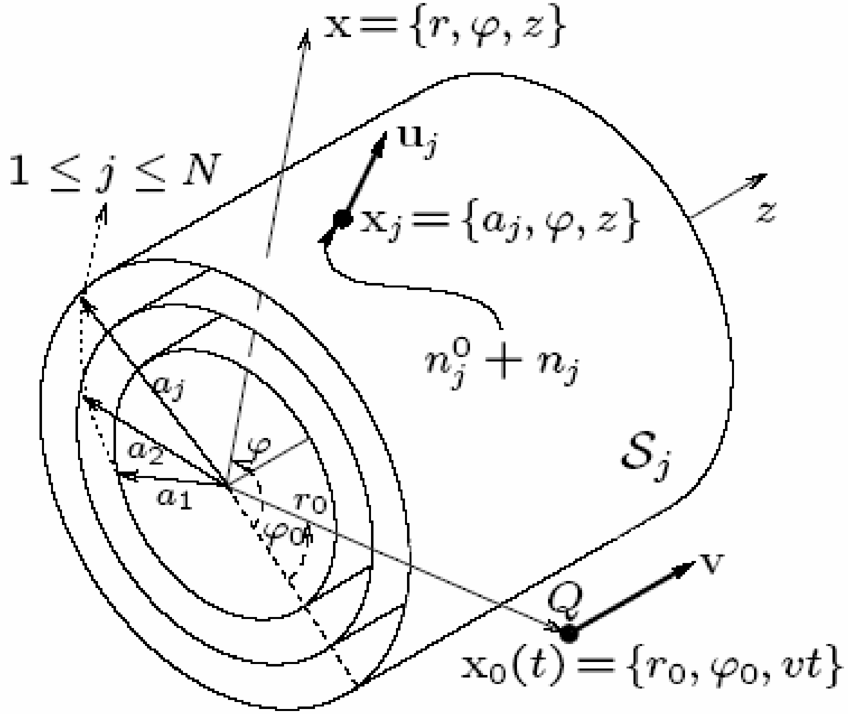
Coulomb explosions during H_2^+ channelling in SWNT

D.P. Zhou *et al.*, PRA *73* (2006) 33202



2D hydrodynamic model of nanotube dielectric response

D.J. Mowbray *et al.,* PRB 70 (2004) 195418



$$\frac{\partial n_j(\mathbf{x}_j,t)}{\partial t} = -n_j^0 \nabla_j \cdot \mathbf{u}_j(\mathbf{x}_j,t)$$

$$\frac{\partial \mathbf{u}_j(\mathbf{x}_j,t)}{\partial t} = \nabla_j \Phi(\mathbf{x},t)|_{r=a_j} - \frac{\alpha_j}{n_j^0}\nabla_j\, n_j(\mathbf{x}_j,t) + \frac{\beta}{n_j^0}\nabla_j\left[\nabla_j^2\, n_j(\mathbf{x}_j,t)\right] - \gamma_j \mathbf{u}_j(\mathbf{x}_j,t)$$

$$\Phi(\mathbf{x},t) = \frac{Q}{\|\mathbf{x}-\mathbf{x}_0(t)\|} - \sum_j \int_{\mathcal{S}_j} d^2\mathbf{x}'_j \, \frac{n_j(\mathbf{x}'_j,t)}{\|\mathbf{x}-\mathbf{x}'_j\|}$$

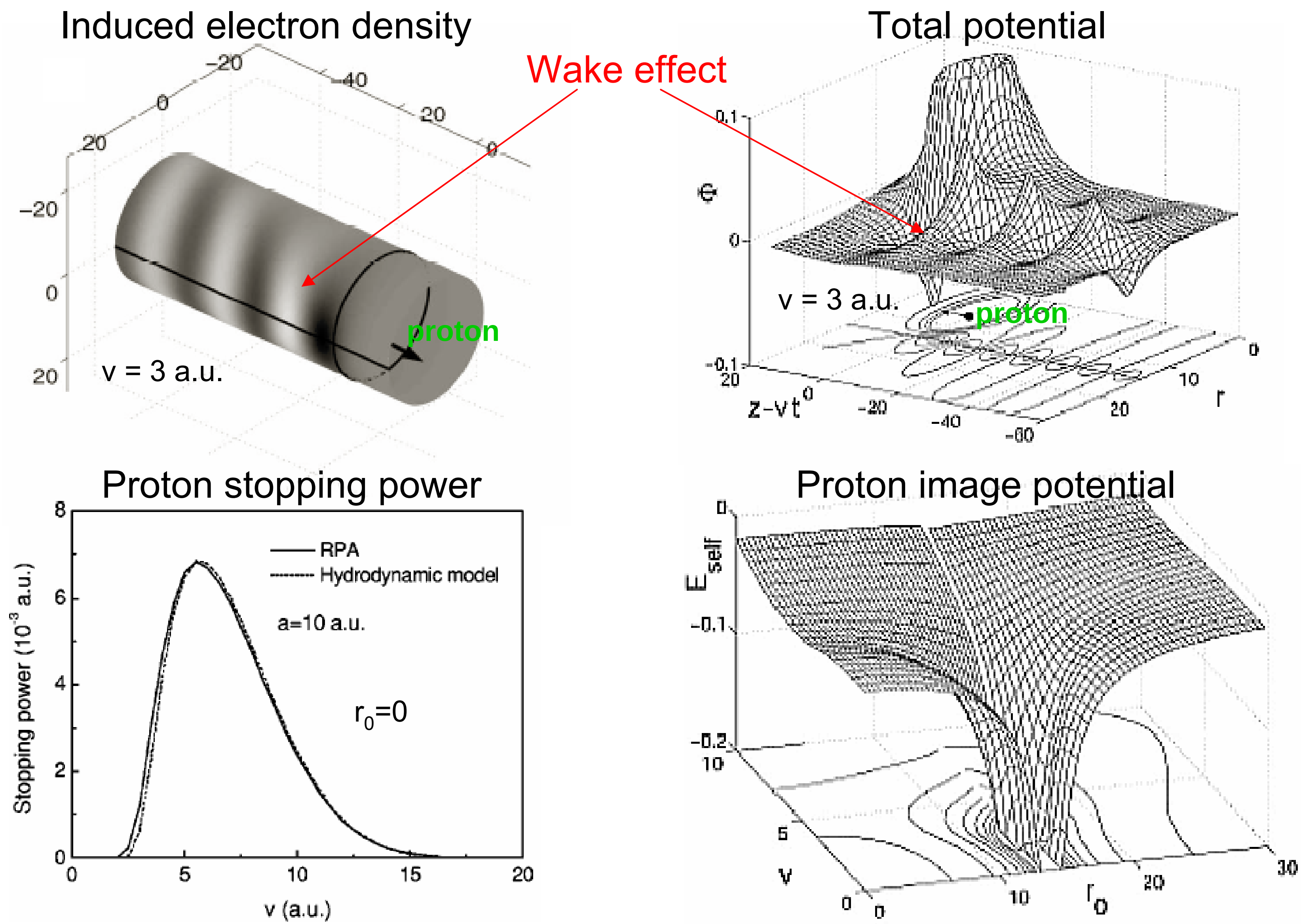
Stopping power

$$S = Q\frac{\partial \Phi_{ind}}{\partial z}\Big|_{\mathbf{x}=\mathbf{x}_0(t)}$$

Self-energy (image potential)

$$E_s = -\frac{Q}{2}\Phi_{ind}\Big|_{\mathbf{x}=\mathbf{x}_0(t)}$$

Dynamic polarization of electrons on SWNT by proton



Coulomb explosions during H_2^+ channelling in SWNT

D.P. Zhou *et al.*, PRA *73* (2006) 33202

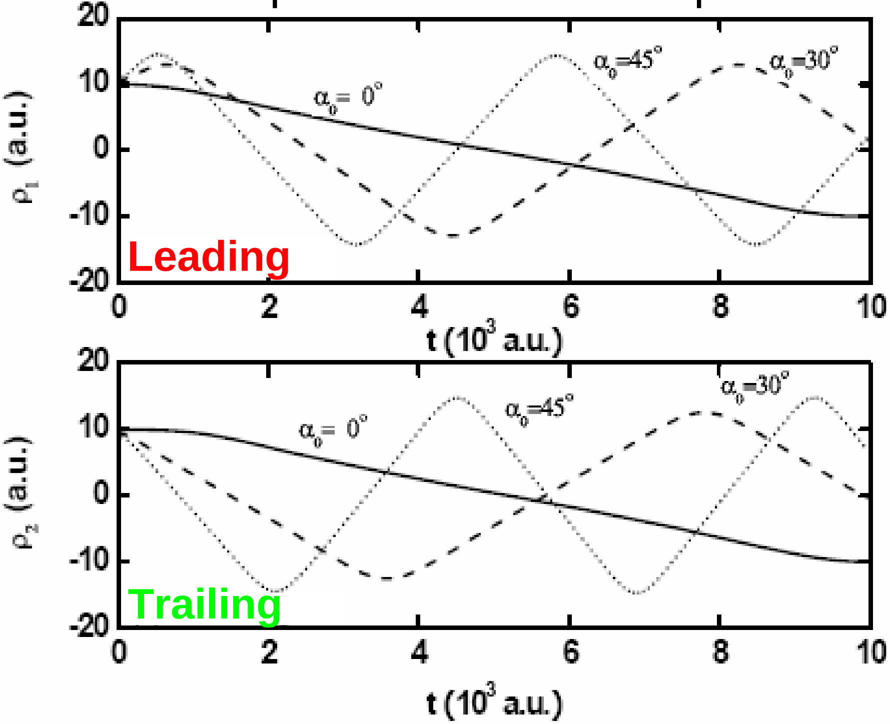
Solve classical equations of motion:

$$\frac{d\mathbf{r}_i}{dt} = \mathbf{u}_i, \qquad M_i \frac{d\mathbf{u}_i}{dt} = \sum_{j(\neq i)=1}^{2} \mathbf{F}_{ij}^{(c)} + \sum_{j=1}^{2} \mathbf{F}_{ij}^{(p)} + \mathbf{F}_i^{(n)}$$

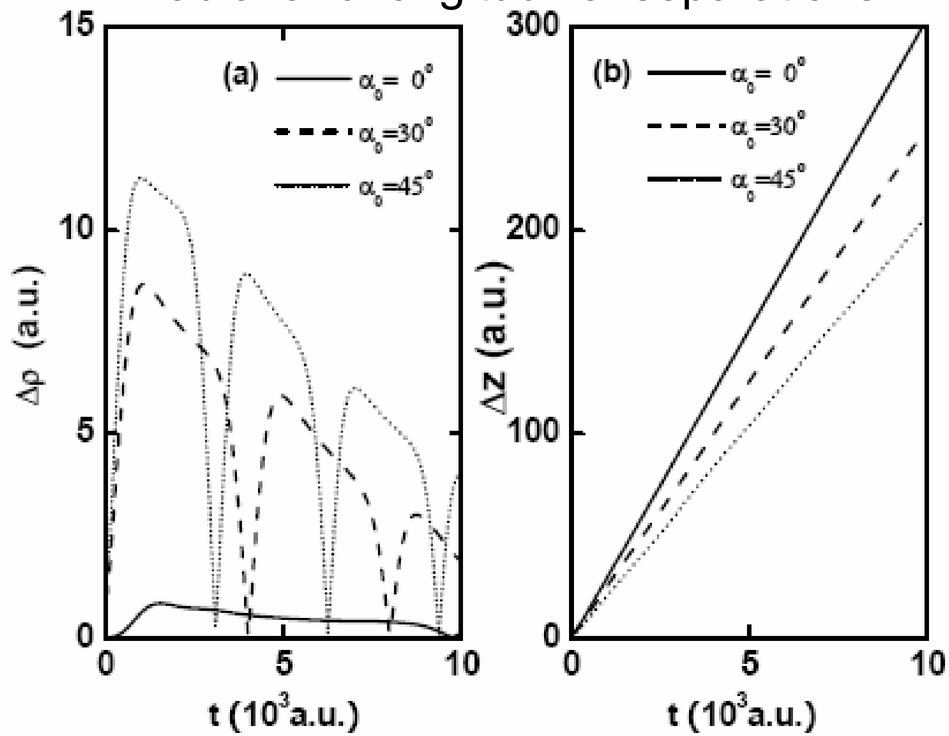
Molecule speed v_0 = 5 a.u. and its alignment angles α_0 = 0°, 30°, 45°

Forces: Coulomb, polarization, Moliere

Radial positions of the two protons



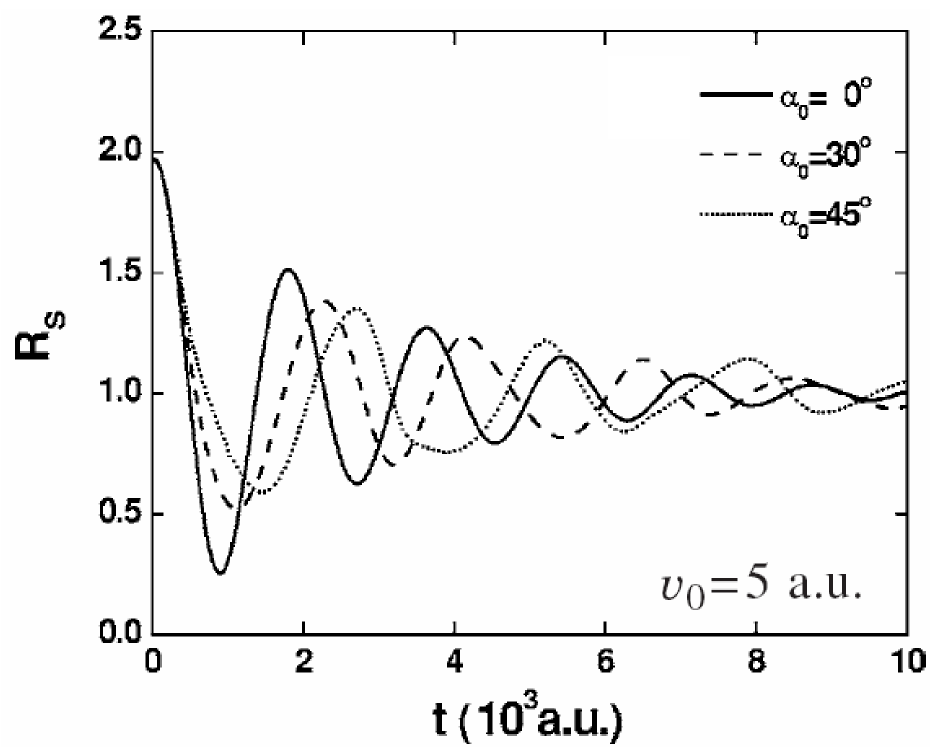
Radial and longitudinal separations



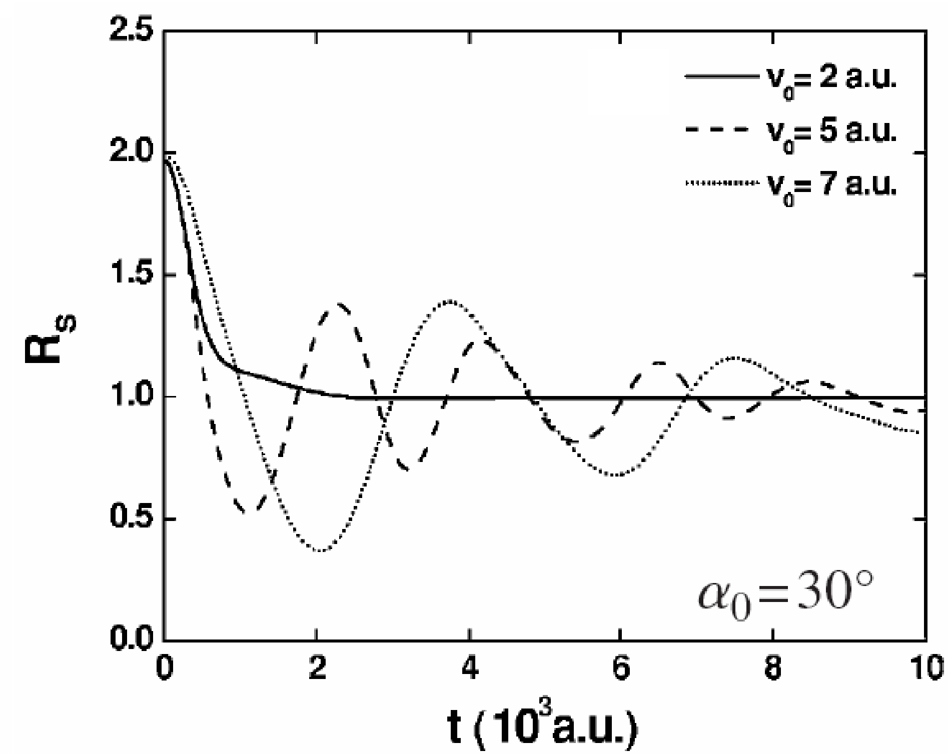
Stopping power ratio vs dwell time for H_2^+ channelling in SWNT

D.P. Zhou *et al.*, PRA *73* (2006) 33202

Effects of alignment angle α_0



Effects of molecule speed v_0



Outlook

❑ Vicinage effects:
- Charge states (demonstrated in both exp. and theory)
- Target polarization effect on Coulomb explosion patterns
- Stopping power (exactly how strong? - still open question)

❑ Experiment: more systematic data on stopping needed

❑ Theory:
- Charge-state modeling
  - ✓ transience effect
  - ✓ exit effect
  - ✓ fluctuations
- Dielectric response models:
  - ✓ inner-shell corrections
  - ✓ non-linearity (Z_1^3 effect)
  - ✓ new targets
- Going beyond dielectric models