Strong-Field Imaging of Molecular Dynamics

Wendell T. Hill, III

University of Maryland
Department of Physics and

Institute for Physical Science and Technology

Explosion Energies

$$E_{ex} = \frac{Z_1 Z_2}{R}$$

Light Atoms

TABLE IV. Total kinetic-energy releases in the fragmentation channels following the multielectron dissociative ionization of N_2, CO, and O_2 in intense laser field with 100-fs pulse duration, 615-nm wavelength in the 10^{15} W/cm^2 range.

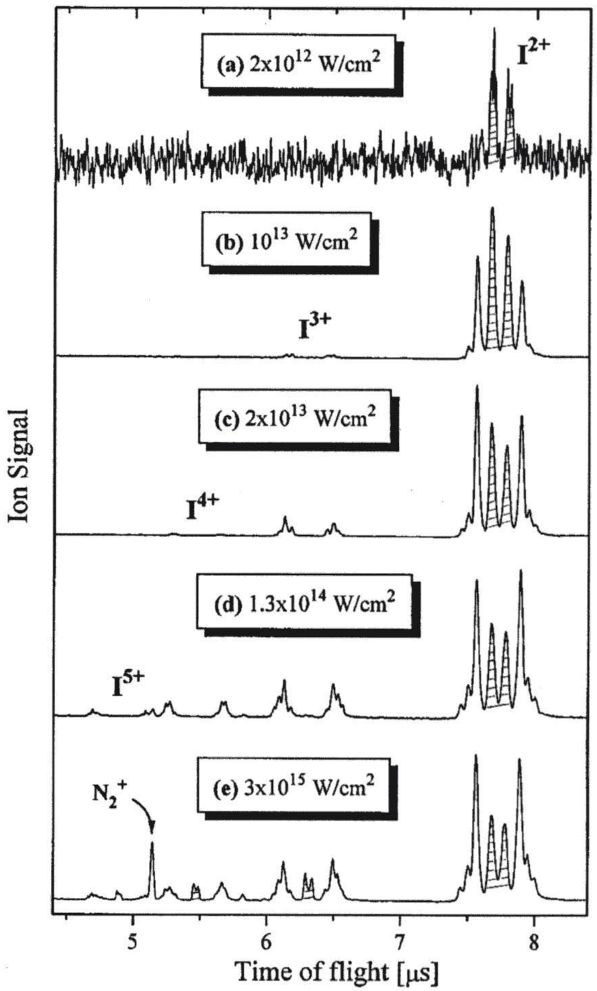
Fragment channel \ Molecule *AB*	N_2	CO	O_2
$A^{+}+B^{+}$	6.8	6.6	5.5
$A^{+}+B^{2+}$		11.7	
$A^{2+}+B^{+}$	13.1	11.7	11.9
$A^{2+}+B^{2+}$	22	19.9	20
$A^{2+}+B^{3+}$		32.7	
$A^{3+}+B^{2+}$	32	29.8	32

Cornaggia, *et al.*, PRA **44**, 4499 (1991)

$R_c \approx 0.28$ nm

~40% of E_{eq}

Heavy Atoms

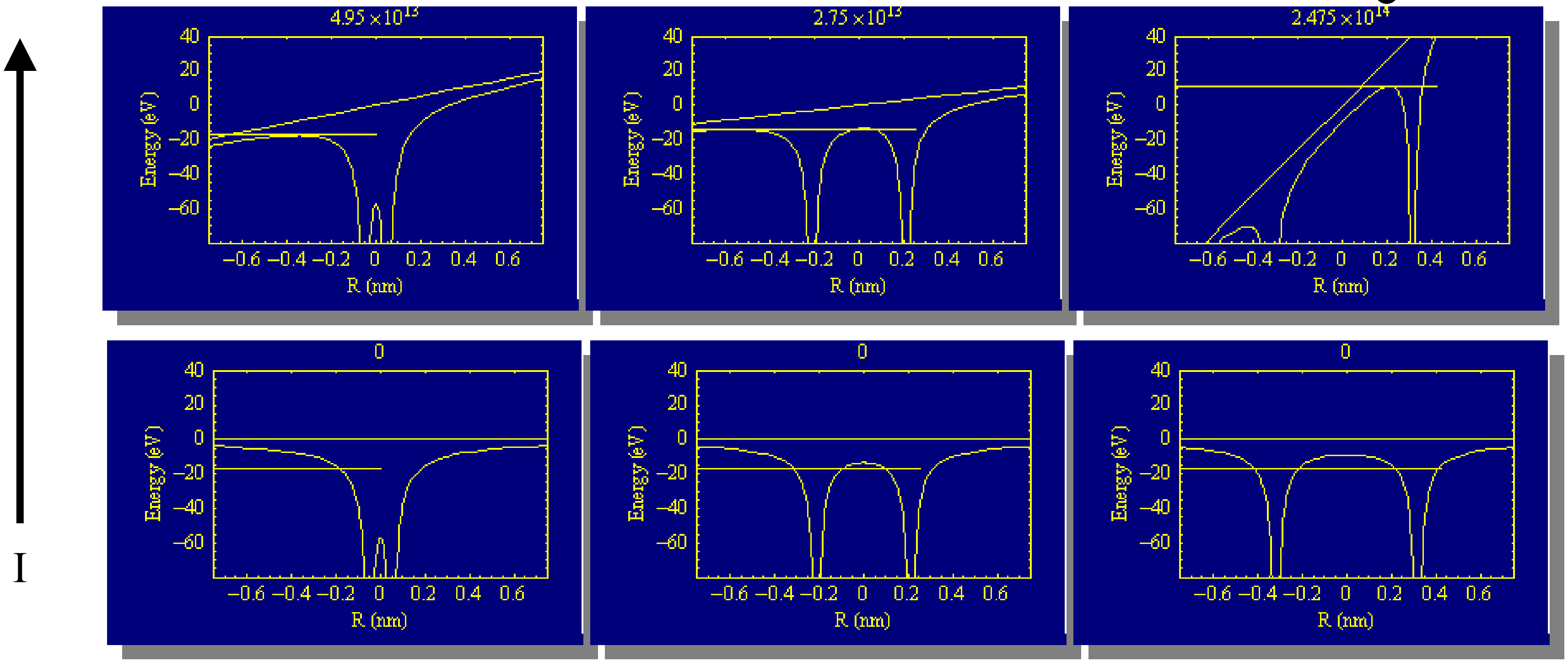


Normand, *et al.*, PRA **53**, R1958 (1996)

$R_c \approx 0.38$ nm

~70% of E_{eq}

Enhanced Ionization at R_c



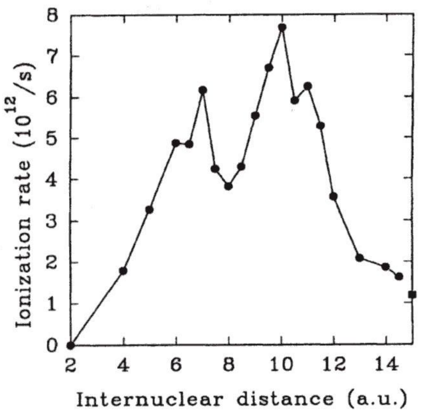
$R = R_{eq}$ $R = R_{eq}$ $R > R_{eq}$

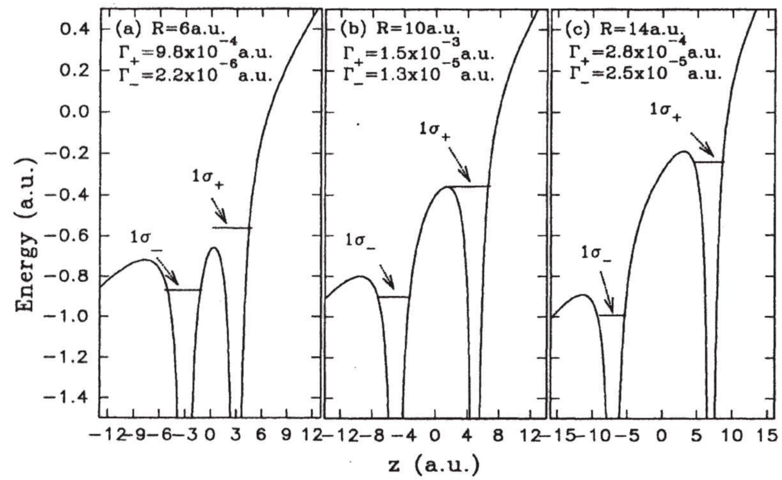
Posthumus, *et al*. J.Phys.**28**; Zuo & Bandrauk, PRA **52**; Seideman , *et al*. PRL **75**

$$1\sigma_g \rightarrow (1s_1 + 1s_2)/\sqrt{2}$$

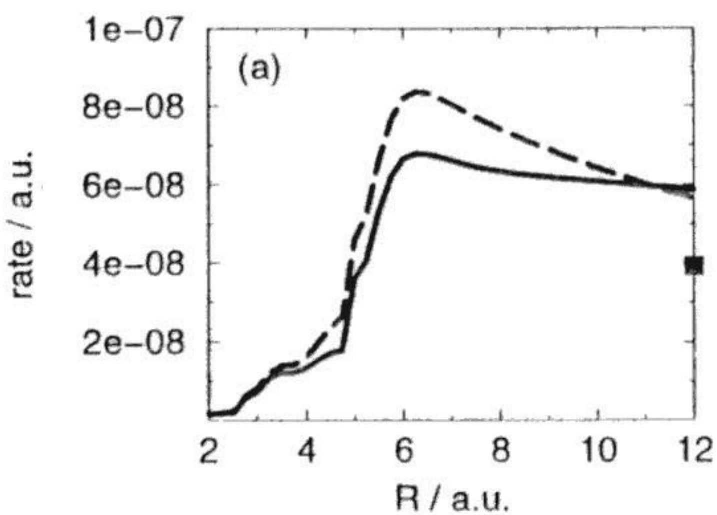
$$1\sigma_u \rightarrow (1s_1 - 1s_2)/\sqrt{2}$$

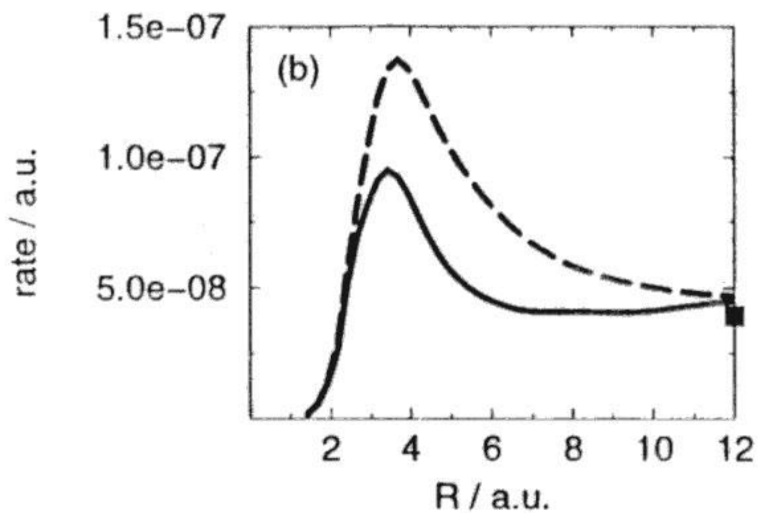
Enhanced Ionization: Theory





Numerical Integration of SE in 3D; Zuo and Bandrauk, PRA 52, R2511 (195)





Manybody S-Matrix; J. Muth-Böhm, A. Becker and F. H. M. Faisal, ICOMP VIII (99)

H_2 Explosion Energy: Experiment

Theory → $R_c=4/I_P$ (au)

Zuo and Bandrauk, PRA **52**, R2511 (1995)

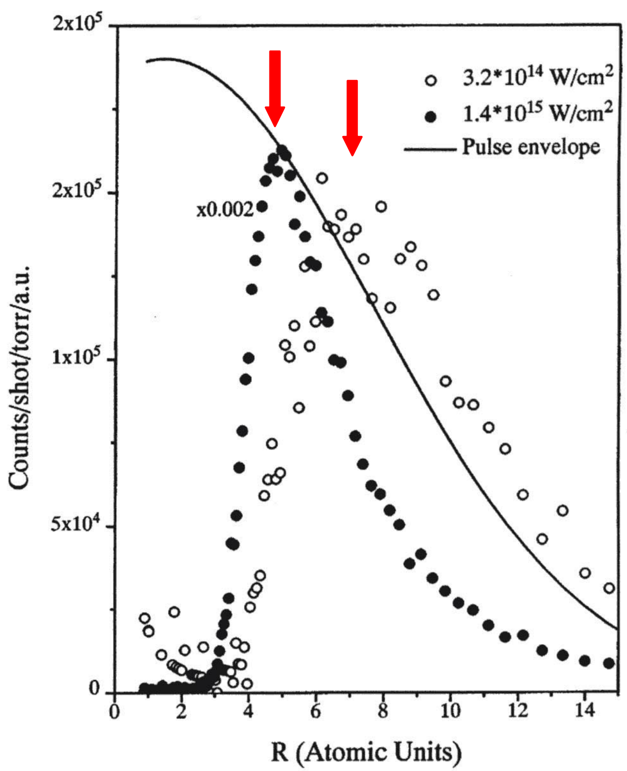
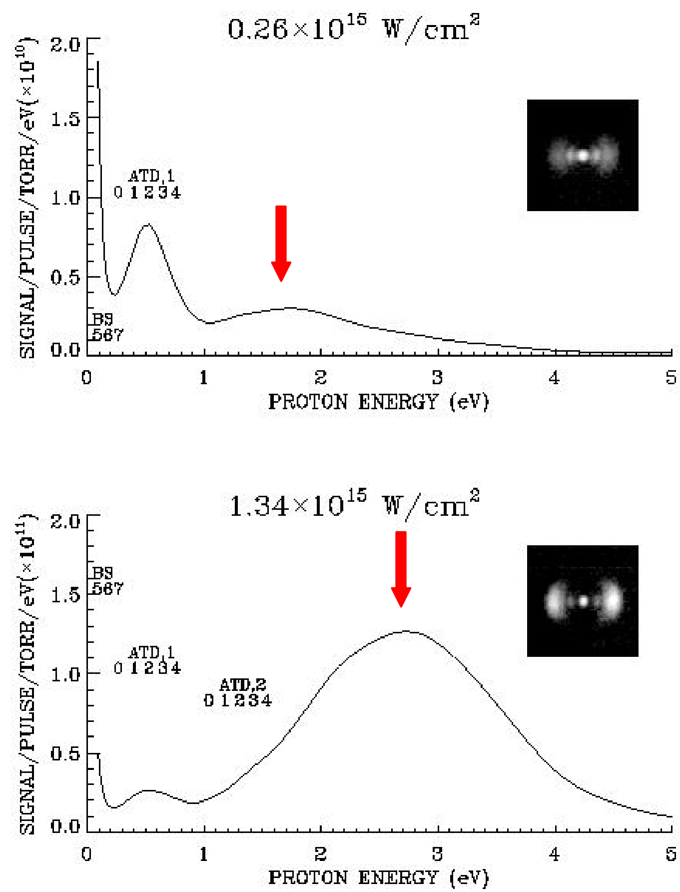


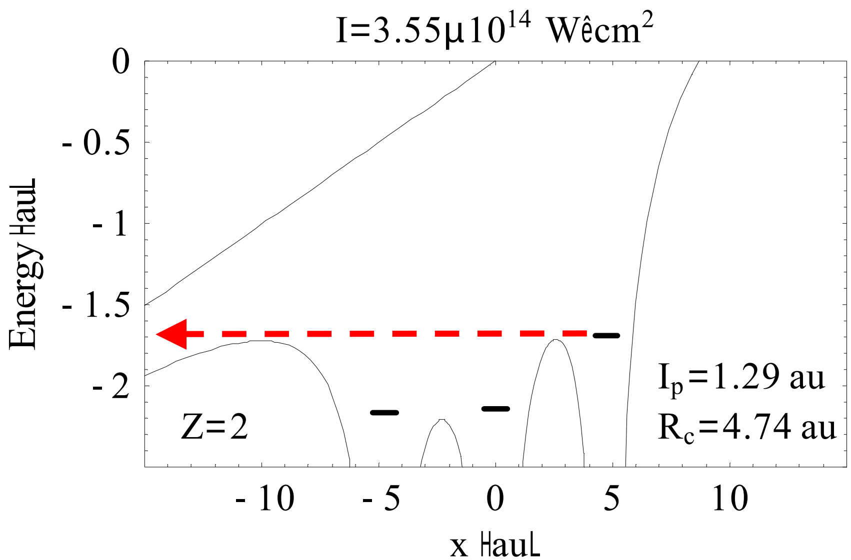
FIG. 4. Ionization signal as a function of internuclear separation and the laser intensity envelope.

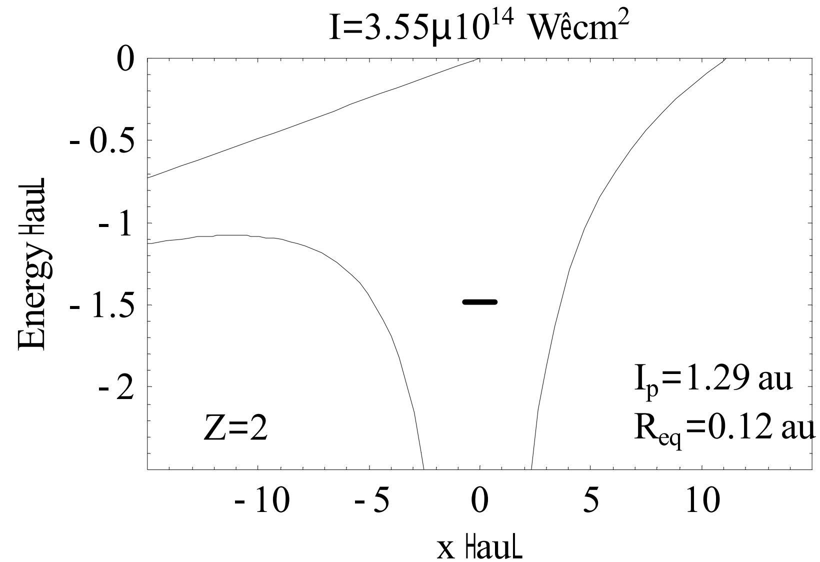
Gibson, *et al*., PRL **79**, 2022 (1997)



Zhang and Hill, unpublished

3-Atom Enhanced Ionization at R_c





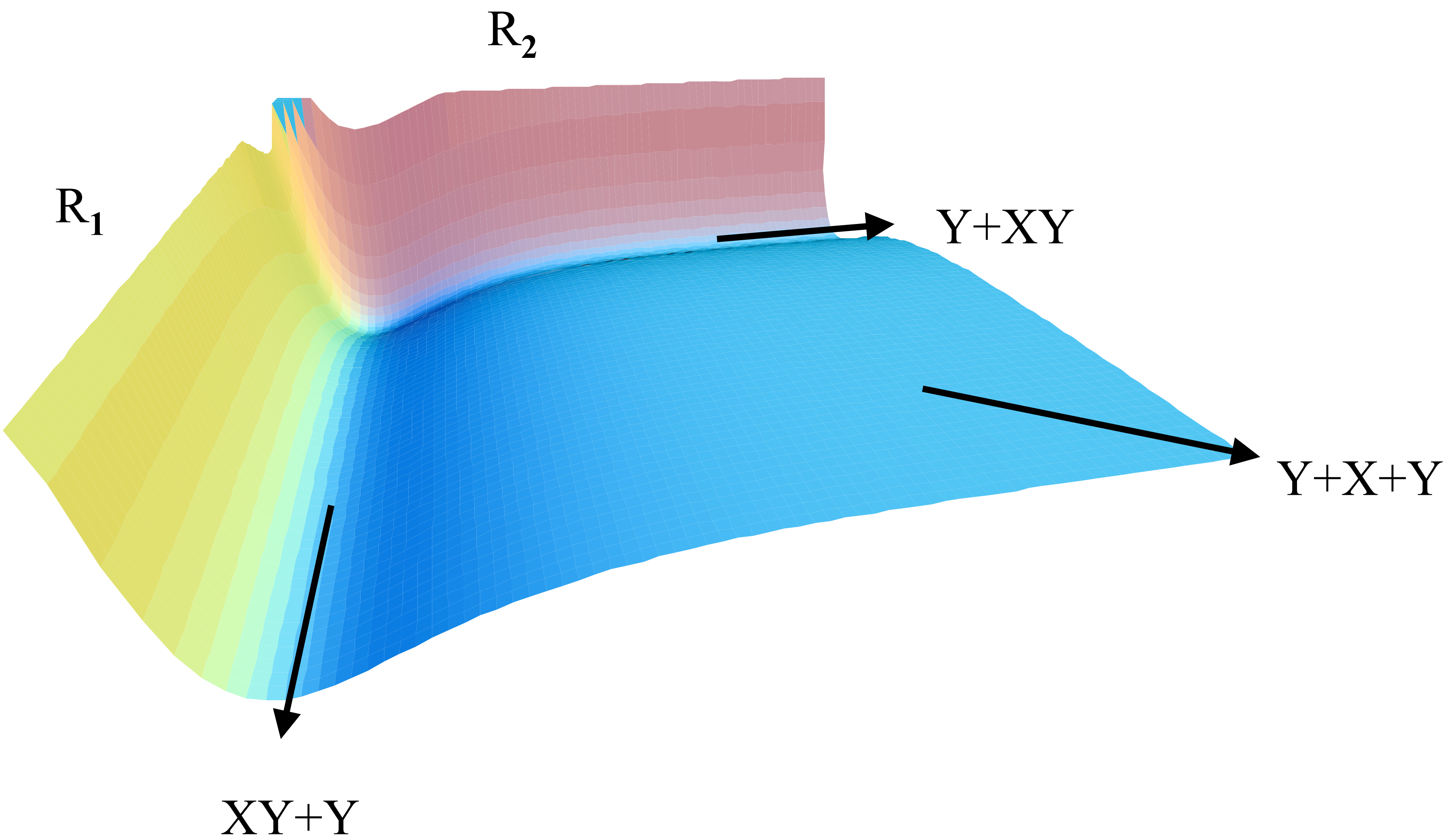
$E_{\text{mea}} \approx 66\ eV$ ➜ $V(r_{CO} = R_C) = \sum_{i<j} \frac{Z_i Z_j}{r_{ij}}$ ➜ $R_C \approx 0.22$ nm (4 au)

$nh\nu + CO_2 \rightarrow O^{p+} + C^{q+} + O^{r+}$

p, q, r = 1,2,3

No intermediate molecular states observed!

GENERIC XY_2 LINEAR POTENTIAL SURFACE



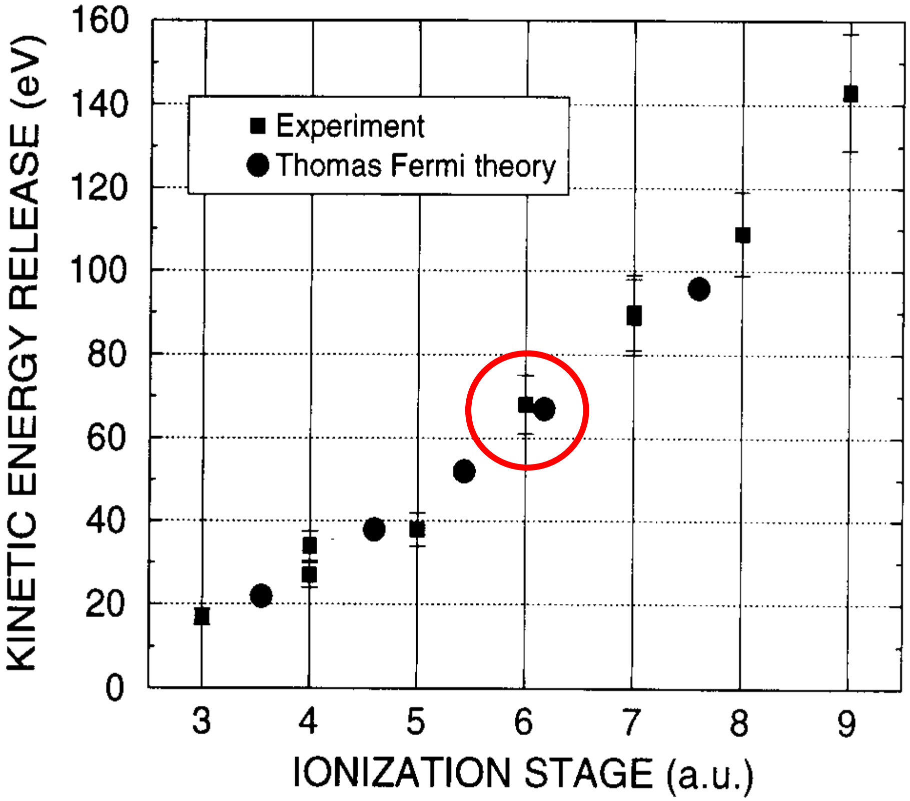
What about the electrons?

Dynamic Screening Explosion Model

Brewczyk, Rzążewski and Clark PRL **78,** 191 (1997)

Ionization and
Dissociation *Interlaced*
Electrons do not leave region immediately!

Dissociation begins @ R_{eq} but slowed by electron cloud producing energies consistent with dissociation from R_c.



Hering, Brewczyk and Cornaggia, PRL **85**, 2288 (2000)

Thomas-Fermi Hydrodynamic Model

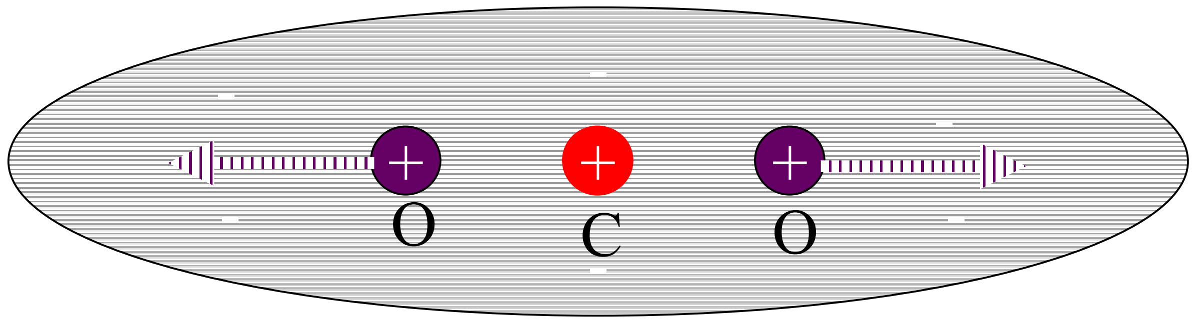
$$\Phi(x,y) = \frac{Ze}{[b^2 + (x-R)^2 + y^2]^{1/2}} + \frac{Z_0 e}{[b^2 + x^2 + y^2]^{1/2}} + \frac{Ze}{[b^2 + (x+R)^2 + y^2]^{1/2}}$$

$$- \frac{e^2}{2Am^2} \int\int_{-\infty}^{\infty} \frac{\Phi(x',y')\,dx'\,dy'}{[c^2 + (x-x')^2 + (y-y')^2]^{1/2}},$$

$$U(R) = -\frac{Ze^2}{2m} \int\int_{-\infty}^{\infty} \frac{\rho_{\rm at}^{Z_0}(x,y) + \rho_{\rm at}^{Z}(x-R,y)}{[b^2 + (x+R)^2 + y^2]^{1/2}}\,dx\,dy - \frac{Ze^2}{2m} \int\int_{-\infty}^{\infty} \frac{\rho_{\rm at}^{Z_0}(x,y) + \rho_{\rm at}^{Z}(x+R,y)}{[b^2 + (x-R)^2 + y^2]^{1/2}}\,dx\,dy$$

$$- \frac{Z_0 e^2}{2m} \int\int_{-\infty}^{\infty} \frac{\rho_{\rm at}^{Z}(x+R,y) + \rho_{\rm at}^{Z}(x-R,y)}{(b^2 + x^2 + y^2)^{1/2}}\,dx\,dy + 2\,\frac{ZZ_0 e^2}{R} + \frac{ZZe^2}{2R},$$

Static Screening Model

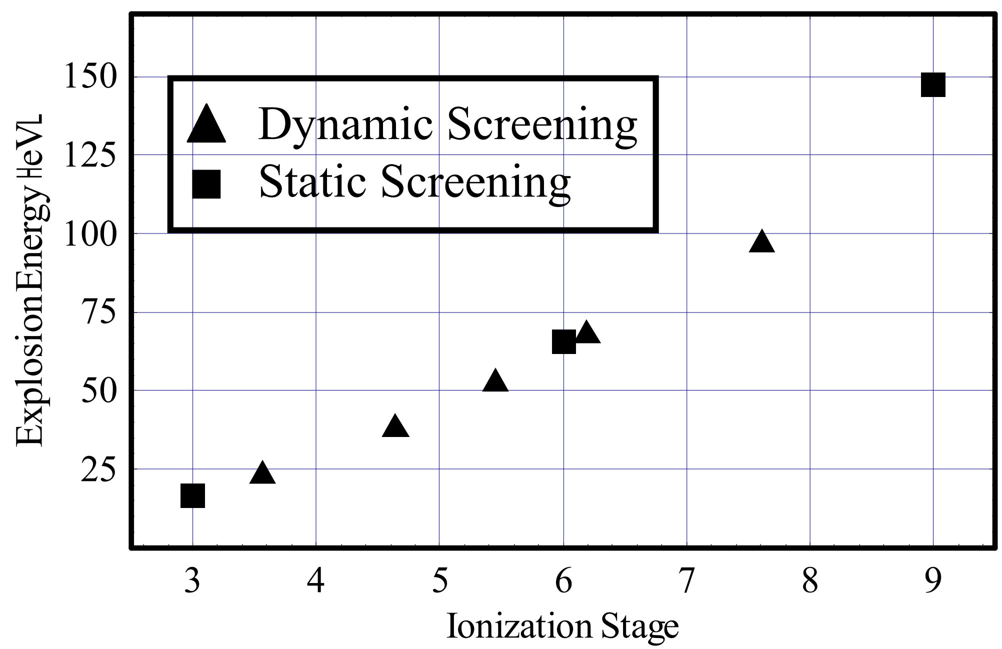


$$Z^{eff} = Z - \sigma$$

$$\sum_{i<j} \frac{Z_i^{eff} Z_j^{eff}}{r_{ij}^{eq}} \approx 66\, e\mathrm{V}$$

$$\sum_{i<j} \frac{Z_i Z_j}{r_{ij}}\bigg|_{r_{CO}=R_C} = \sum_{i<j} \frac{Z_i^{eff} Z_j^{eff}}{r_{ij}}\bigg|_{r_{CO}=R_{eq}}$$

$$\sigma \equiv Z\left(1 - \sqrt{R_{eq}/R_C}\right)$$



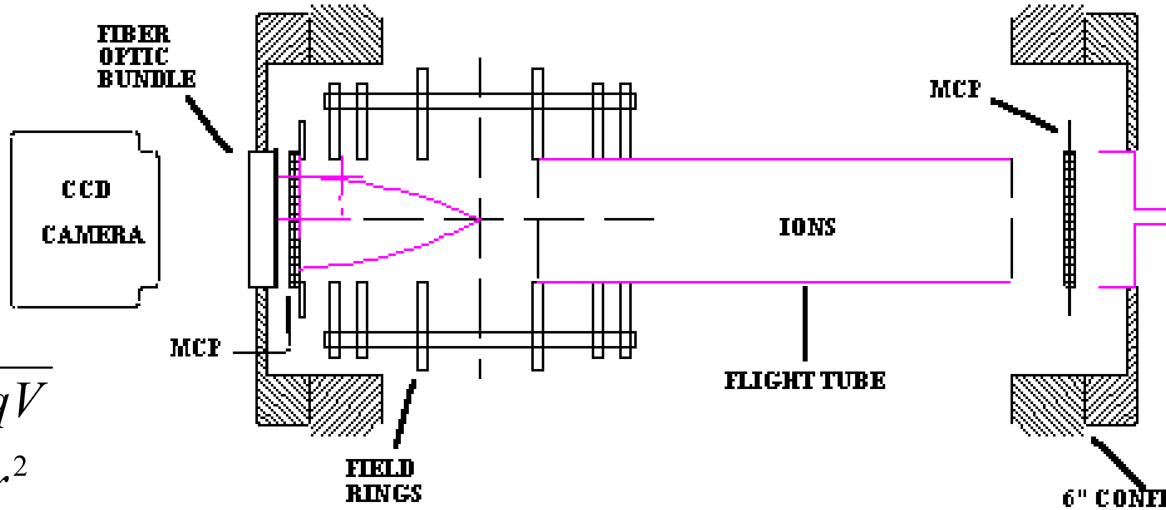
Zhao, Zhang and Hill, PRA (submitted)

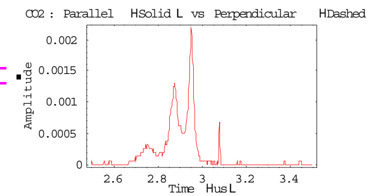
Angular Dependence of E_C

$$\varepsilon = \frac{2Z^2}{R_C(\theta_b)} + \frac{Z^2}{2R_C(\theta_b)\sin(\theta_b/2)}$$

$$\varepsilon = \frac{2\left[Z - \sigma(\theta_b)\right]^2}{R_{eq}} + \frac{\left[Z - \sigma(\theta_b)\right]^2}{2R_{eq}\sin(\theta_b)}$$

Momentum & Time-of-Flight Spectra





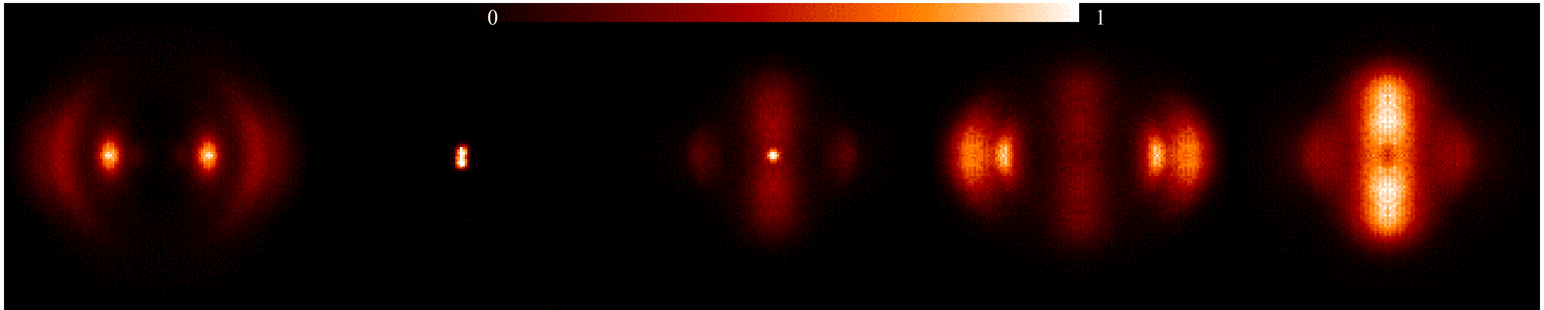
$$p_0 \propto r\sqrt{qV}$$

$$E_0 \propto qVr^2$$

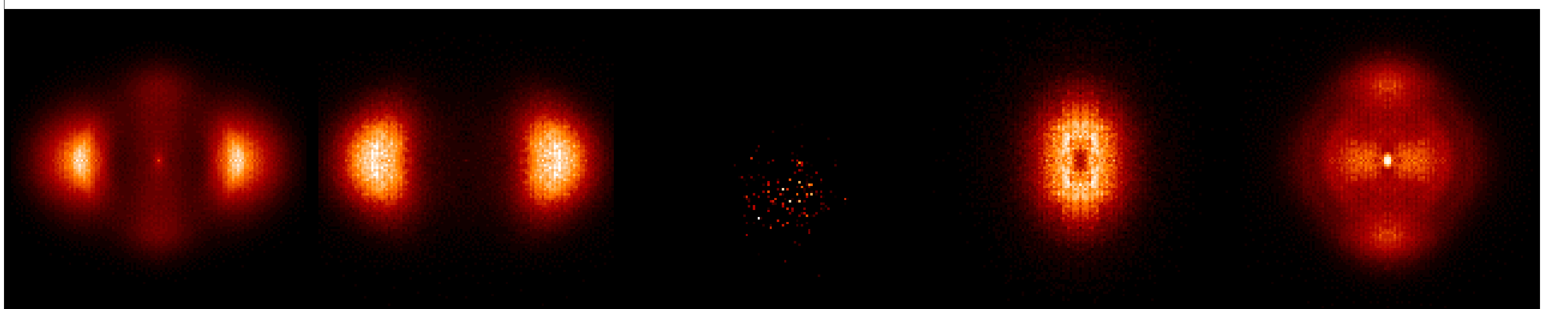
$$E_0 = f(t;m,q)$$

Time-Resolved Images
128 x 128 Pixels
730 Hz Digital Acquisition
500,000 – 1,000,000 Frames
Mass-Resolved
Energies & Angular Distributions

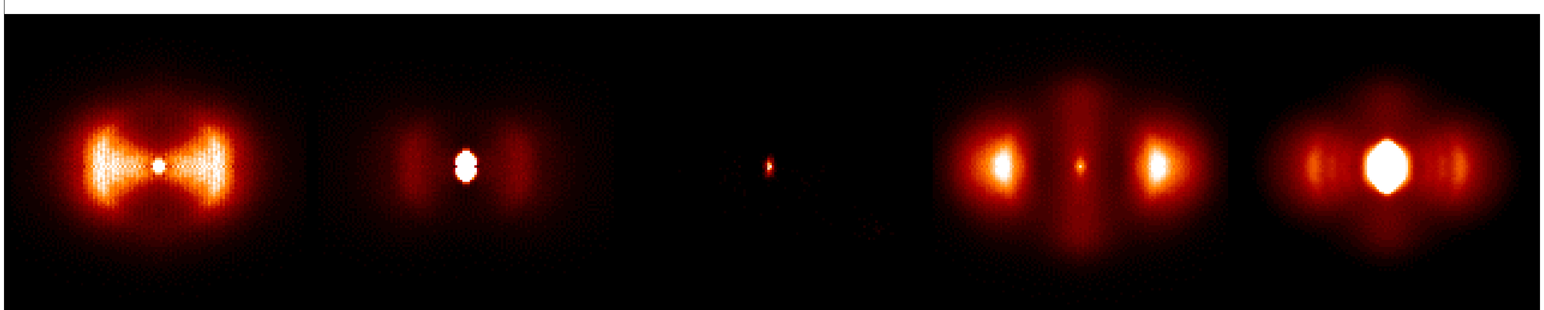
Time-Resolved Waveforms
Digital Scope Acquisition
Mass-Resolved
Energies Distributions



(a) H^+ 110 – 210 ns | (b) H_2^+ 210 – 310 ns | (c) N^{3+} 320 – 420 ns | (d) N^{3+} , O^{3+} 370 – 470 ns | (e) N^{2+} 420 – 520 ns

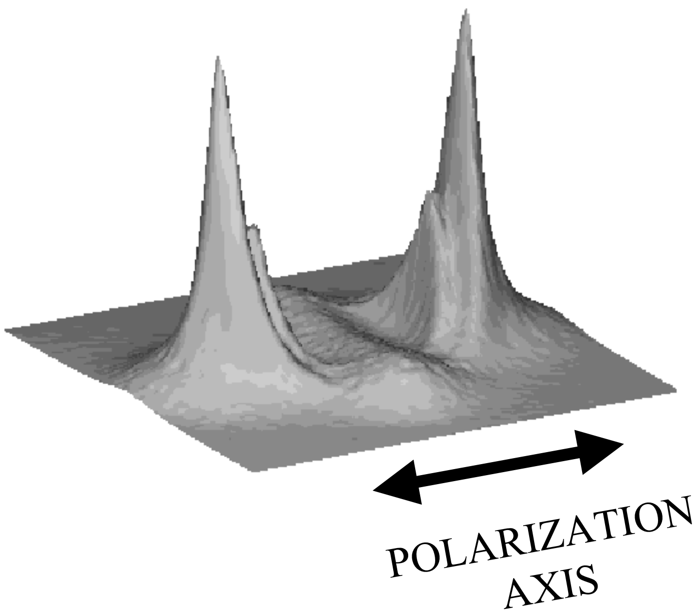


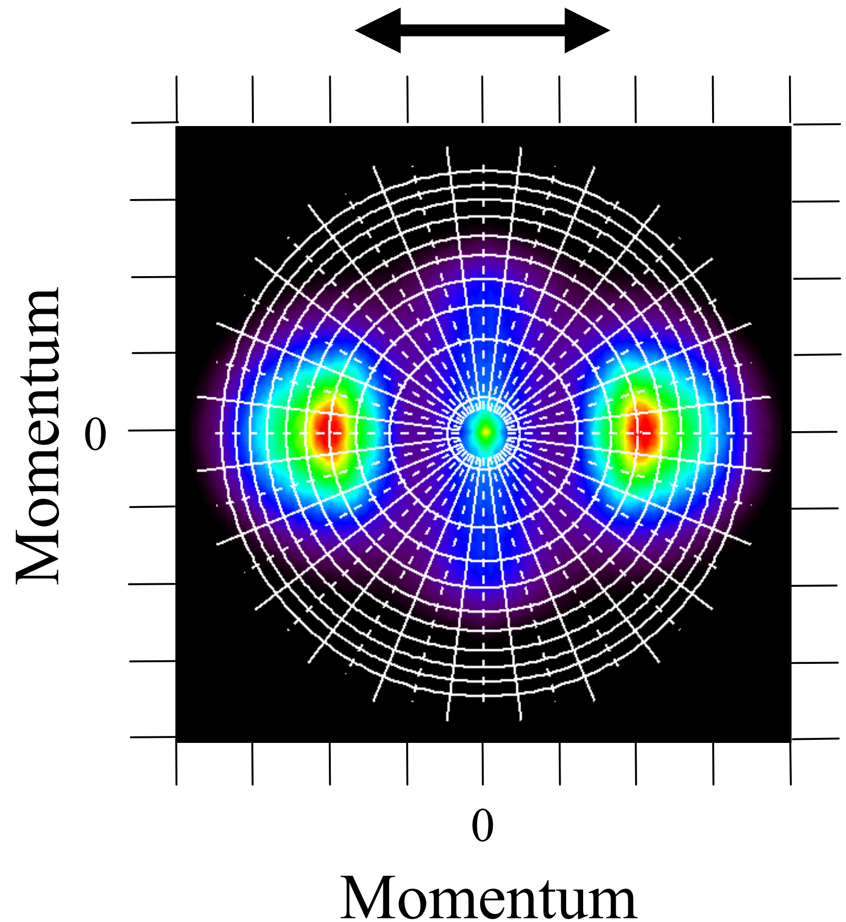
(f) N^{2+} , O^{2+} 470 – 570 ns | (g) O^{2+} 520 – 620 ns | (h) 570 – 670 ns | (i) N^+ 620 – 720 ns | (j) N^+ 670 – 770 ns



(k) O^+ 720 – 820 ns | (l) O^+ 770 – 870 ns | (m) OH^+ 820 – 920 ns | (n) $N^{2+,3+}$, $O^{2+,3+}$ 320 – 620 ns | (o) All Ions All Time

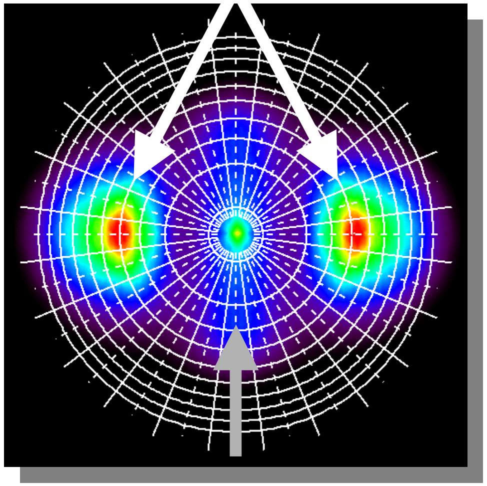
Momentum Image



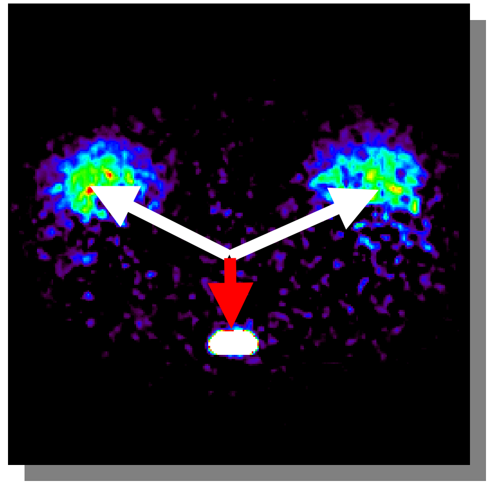


Scale: 5 – 8 amu/div

Correlation Imaging



AVERAGE IMAGE



SELECTIVE AVERAGE

POLARIZATION
AXIS

Image labeling: a graphical interface to correlation in multiparticle ejection dynamics

K. Zhao,[1,2] G. Zhang,[3] and W. T. Hill, III[1,2]

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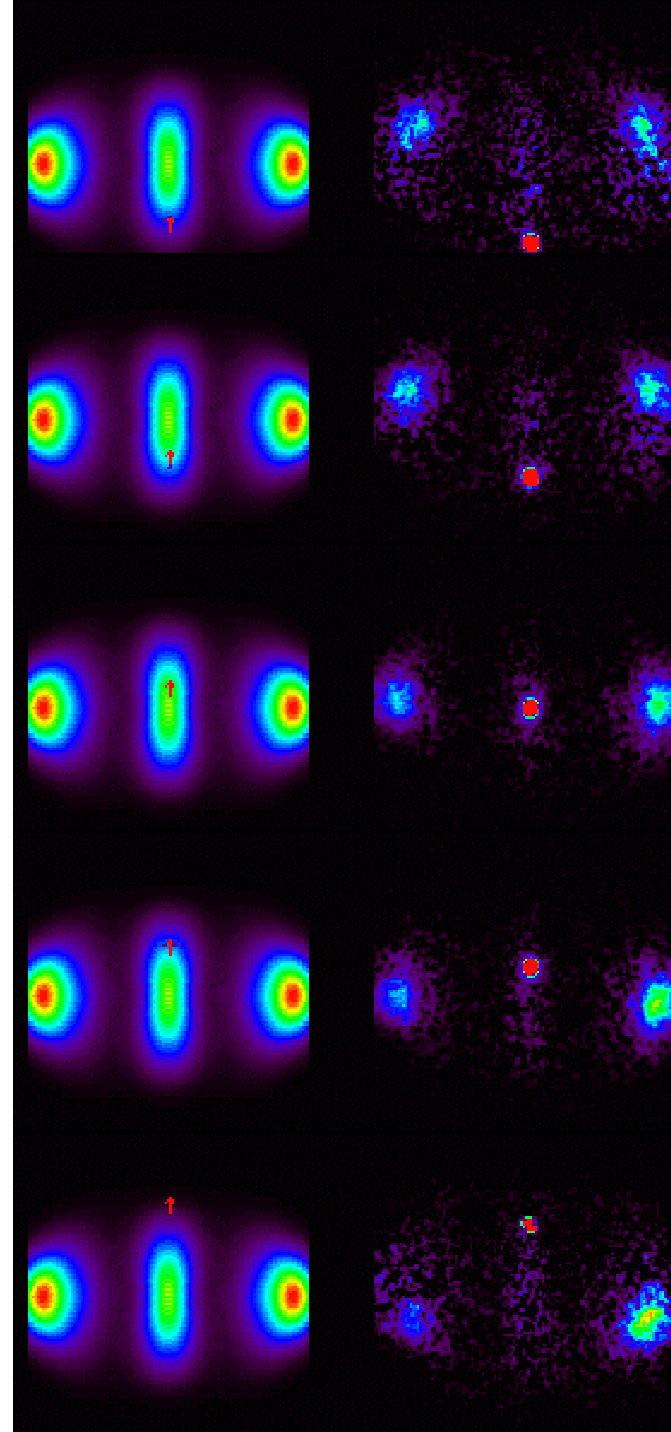
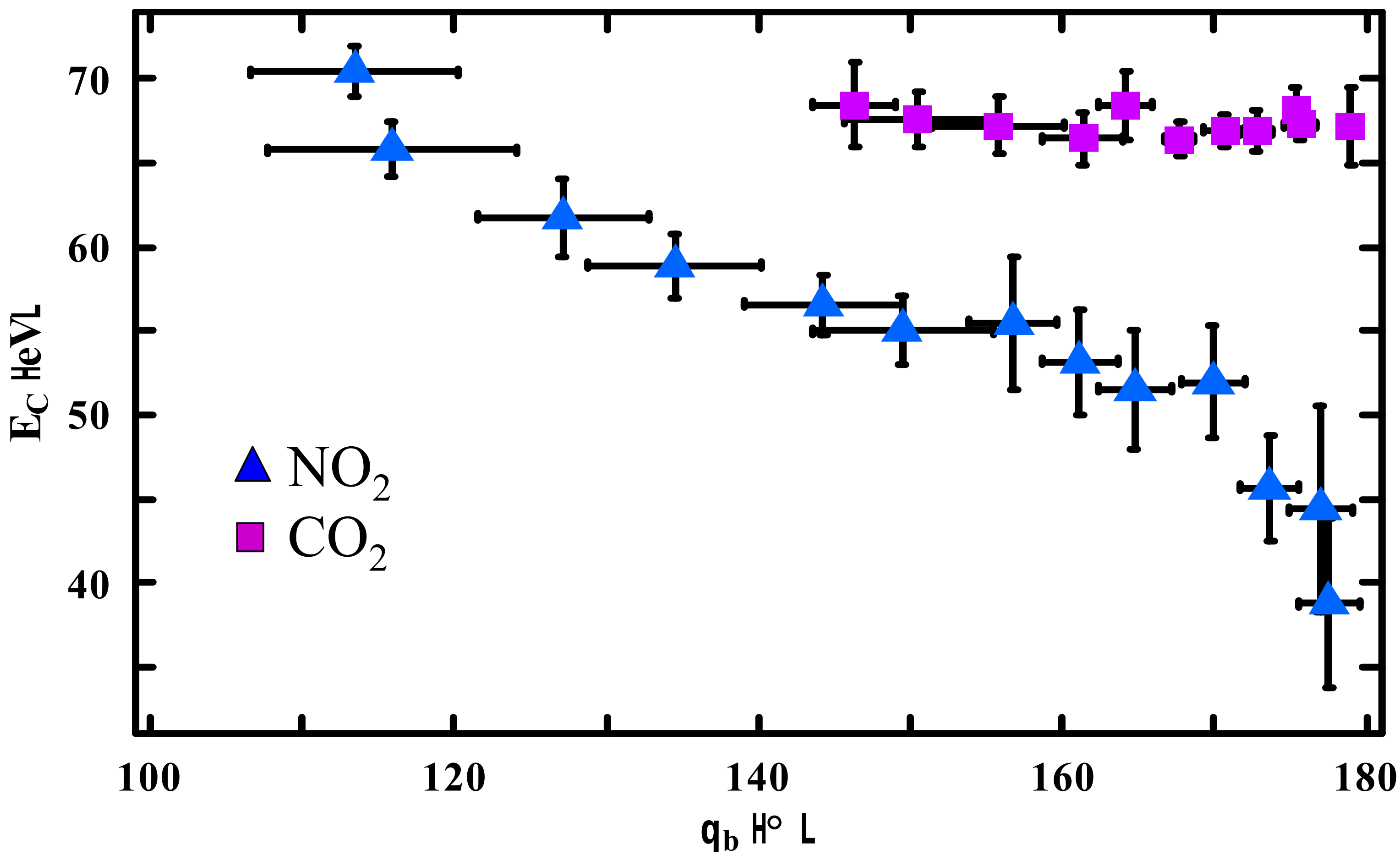
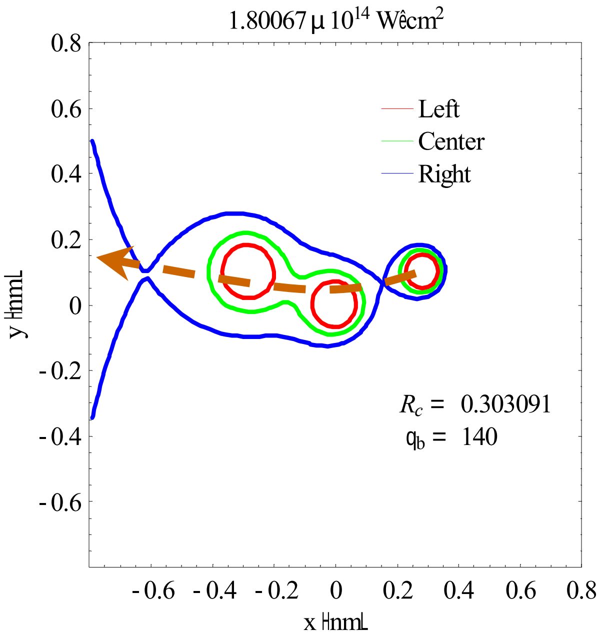


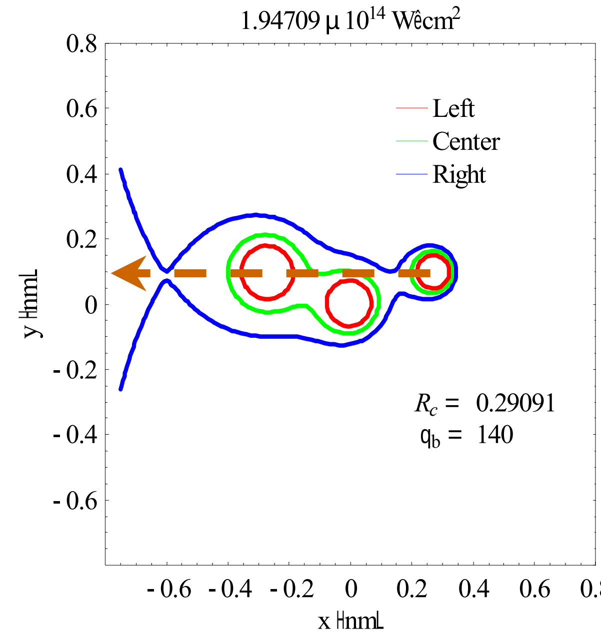
Symmetric Coulomb Explosion



Through Minimum



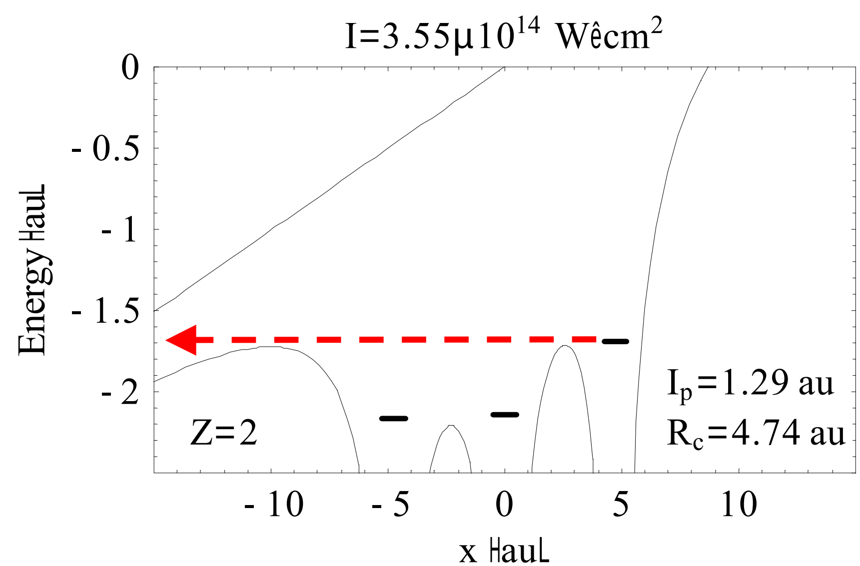
Across O-O



Over-the-Barrier Ionization

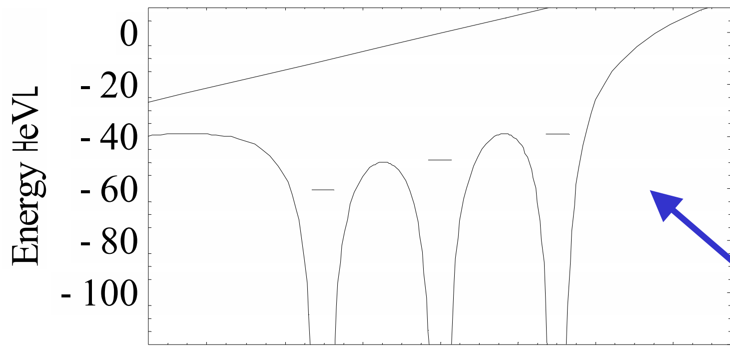
$$E_C = \frac{2Z^2}{R_C(\theta_b)} + \frac{Z^2}{2R_C(\theta_b)\sin(\theta_b/2)}$$

$$V = -\sum_{i=1}^{3} \frac{Z_i}{\sqrt{(x-x_i)^2 + (y-y_i)^2 + s^2}} + Fx$$

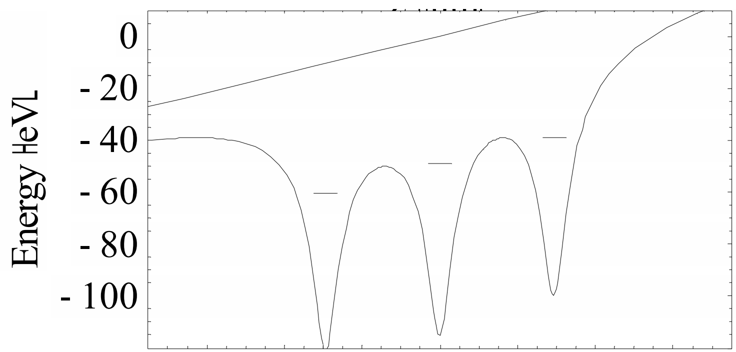


Limits on Smoothing Factor

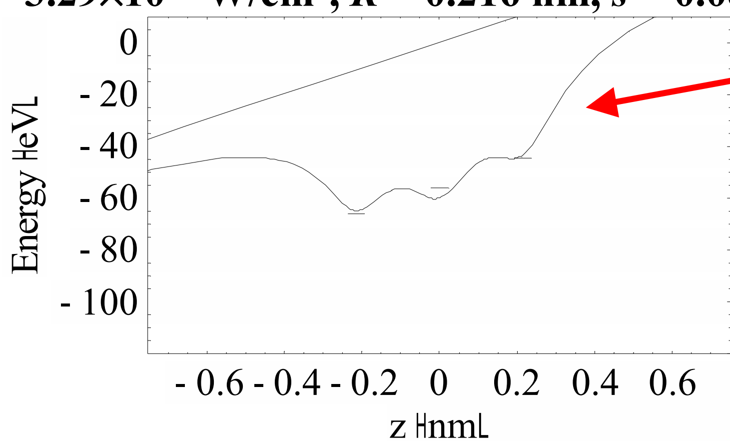
I **= 1.66×10^14 , *R* = 0.302 nm, *s* = 0.0001**

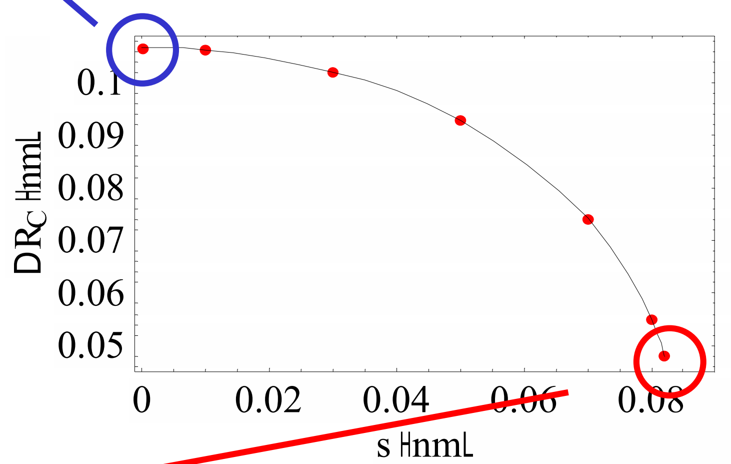


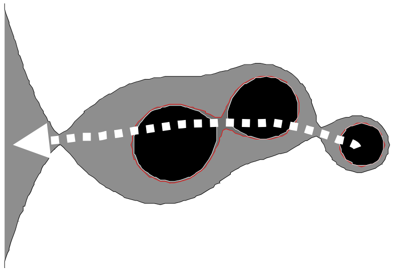
I **= 1.75 ×10^14 W/cm², *R* = 0.294 nm, *s* = 0.03**



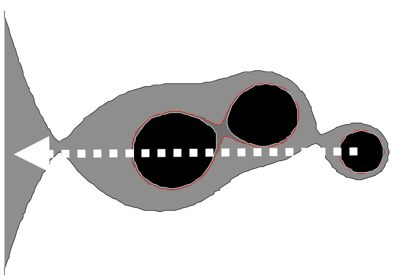
I **= 3.29×10^14 W/cm², *R* = 0.216 nm, *s* = 0.082**

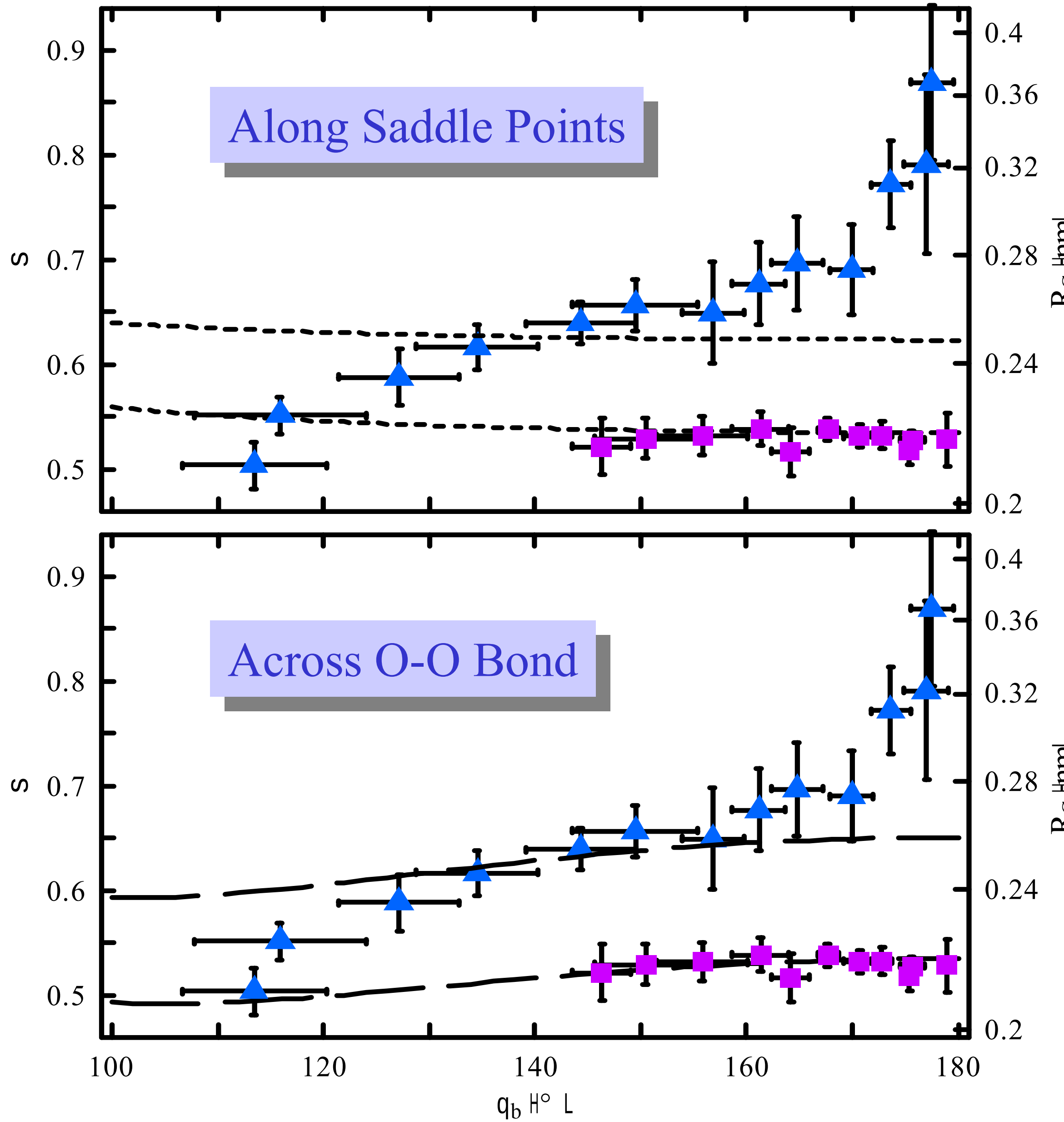




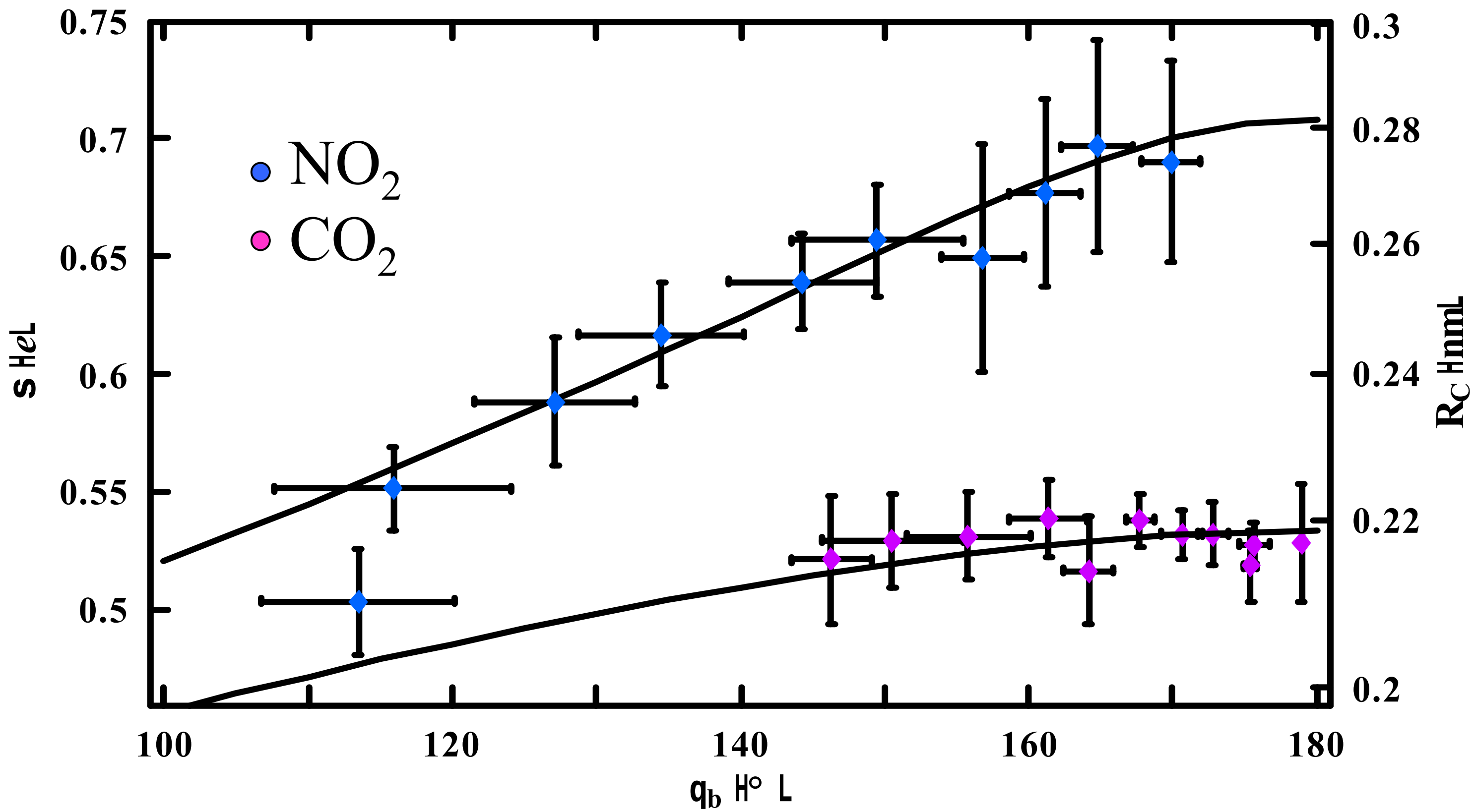


Enhanced Ionization at R_c





Symmetric Explosion vs. Bond Angle



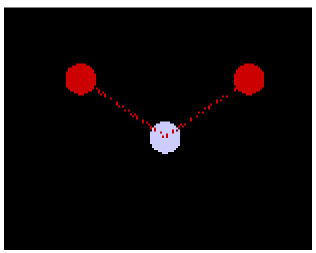
$$V = Z_{eff}^2(\theta_b)\left[\frac{2}{R_{eq}} + \frac{1}{2R_{eq}\sin(\theta_b/2)}\right]$$

$$= Z^2\left[\frac{2}{R_{eq}} + \frac{1}{2R_{eq}\sin(\theta_b/2)}\right] - \sum_{i=1}^{3}\frac{Z_i q_{el}}{\pi}\int_{Area}\frac{\rho(x,y)\,dxdy}{\sqrt{(x-x_i)^2+(y-y_i)^2+s^2}}$$

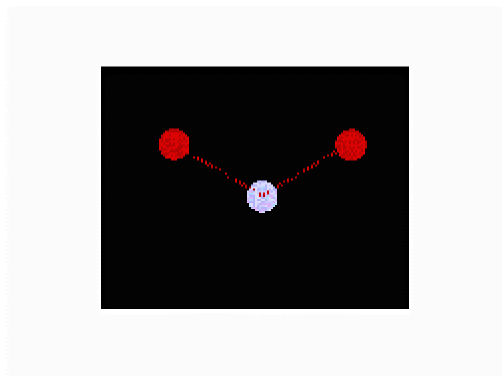
$$= e^{-\frac{x^2}{x_0{}^2}-\frac{y^2}{y_0{}^2}}$$

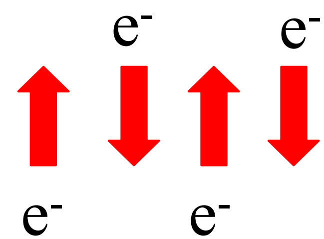
Charge Density Parameters

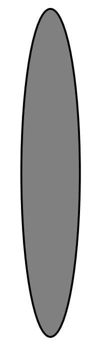
NO_2



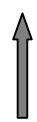
CO_2







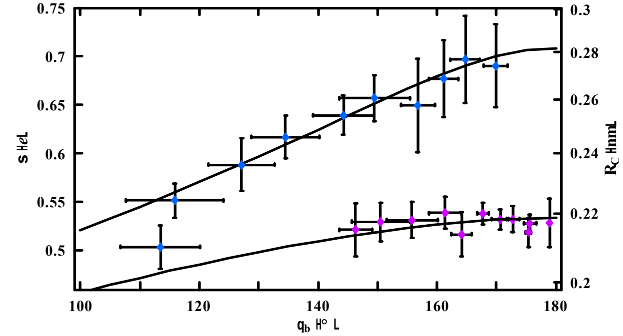
Resulting Charge Distribution

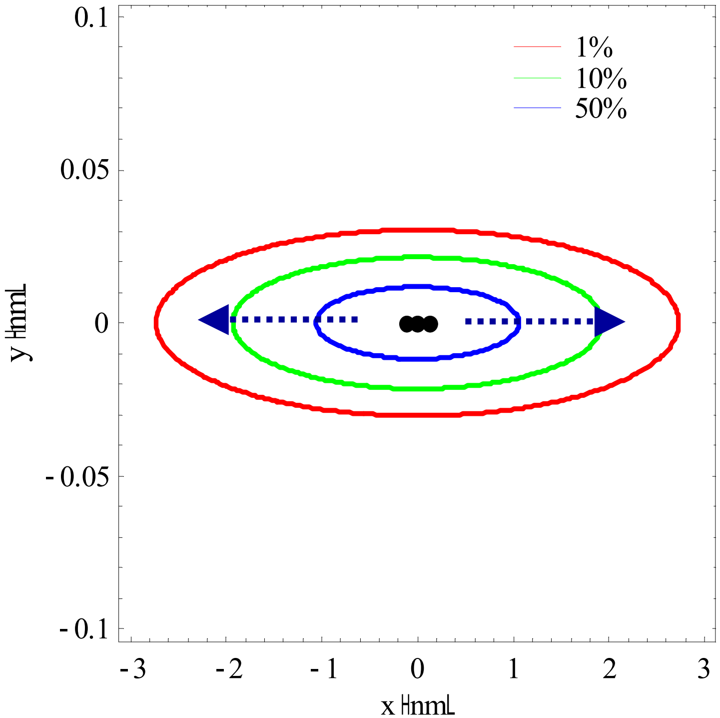


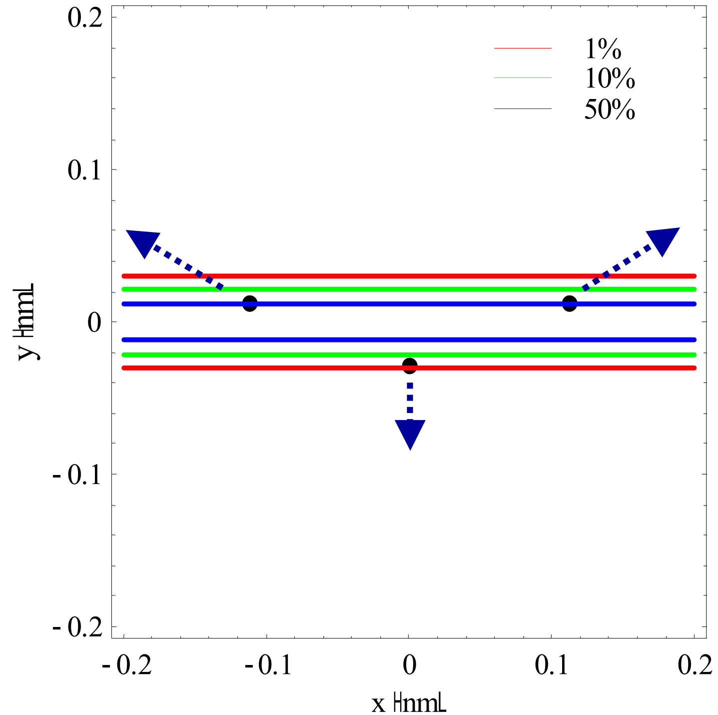
Wider Distribution

Larger y_0

Linear vs. Bent Explosions







CO_2 explosion range
θ_b = 145 - 180°

NO_2 explosion range
θ_b = 125 - 170°

Conclusion

- The first example of where the R_c and screening models give different results!
- Screening explains the electron dynamics and the differences between linear and bent explosions.
- We have now recovered the ability to use ultrafast pulses to examine molecular structure.

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