# Frustrated Coulomb explosion of helium nanodroplets

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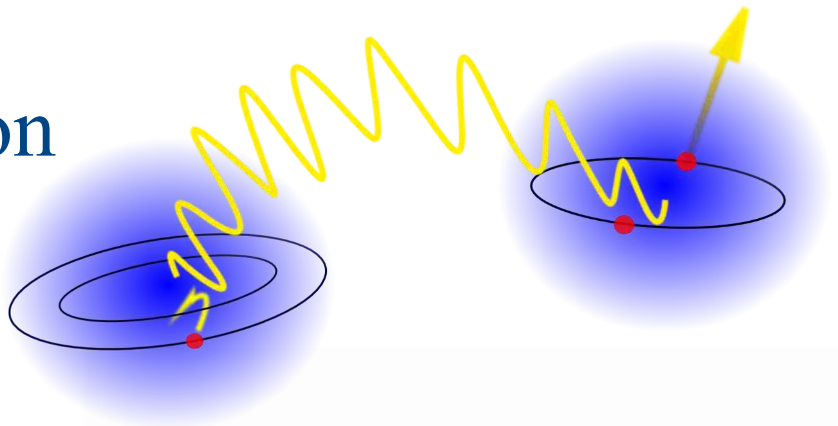
SFP, July 2023

# Outline

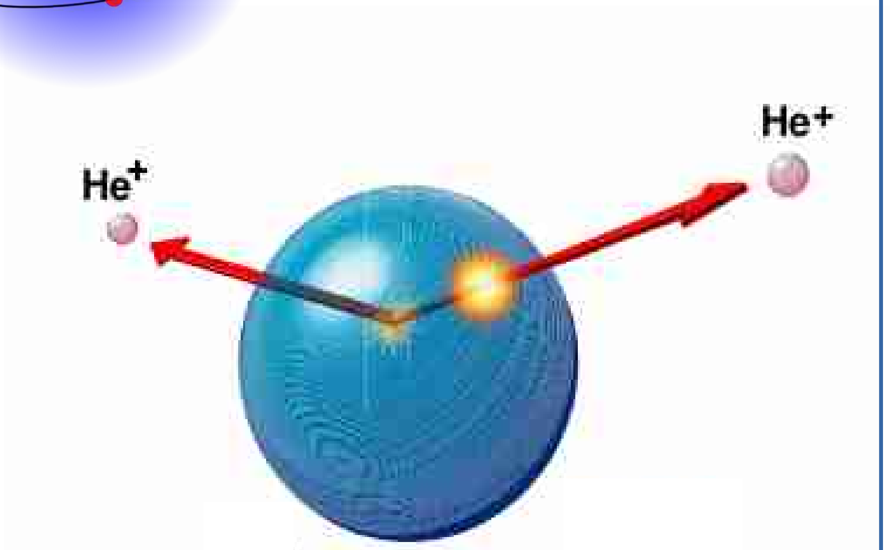
Motivation

Interatomic Coulombic Decay (ICD) in helium clusters



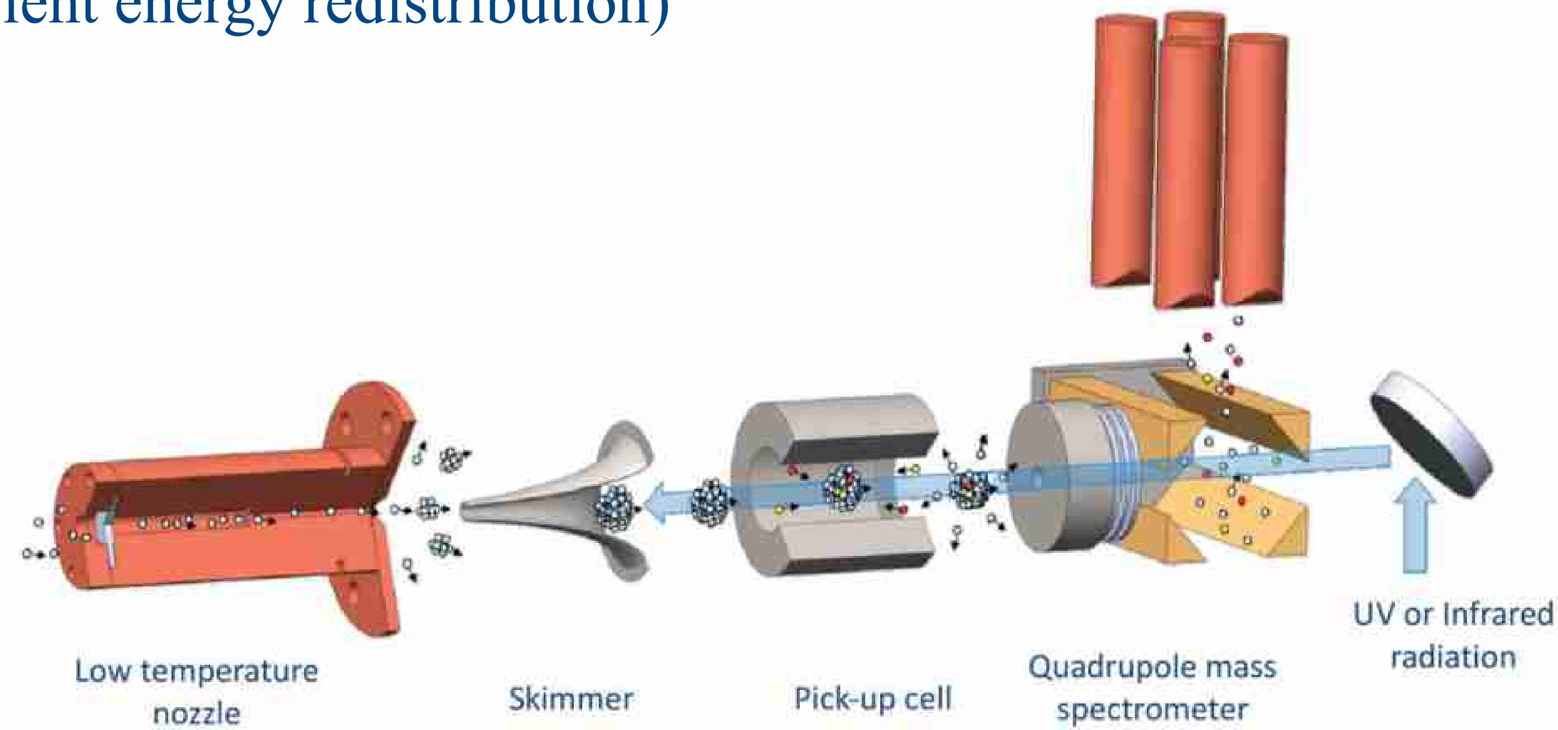
Frustrated Coulomb Explosion

*Theoretical approach*



# Motivation

Cold isothermal nanoscopic reactors
(efficient energy redistribution)




How charges redistribute?

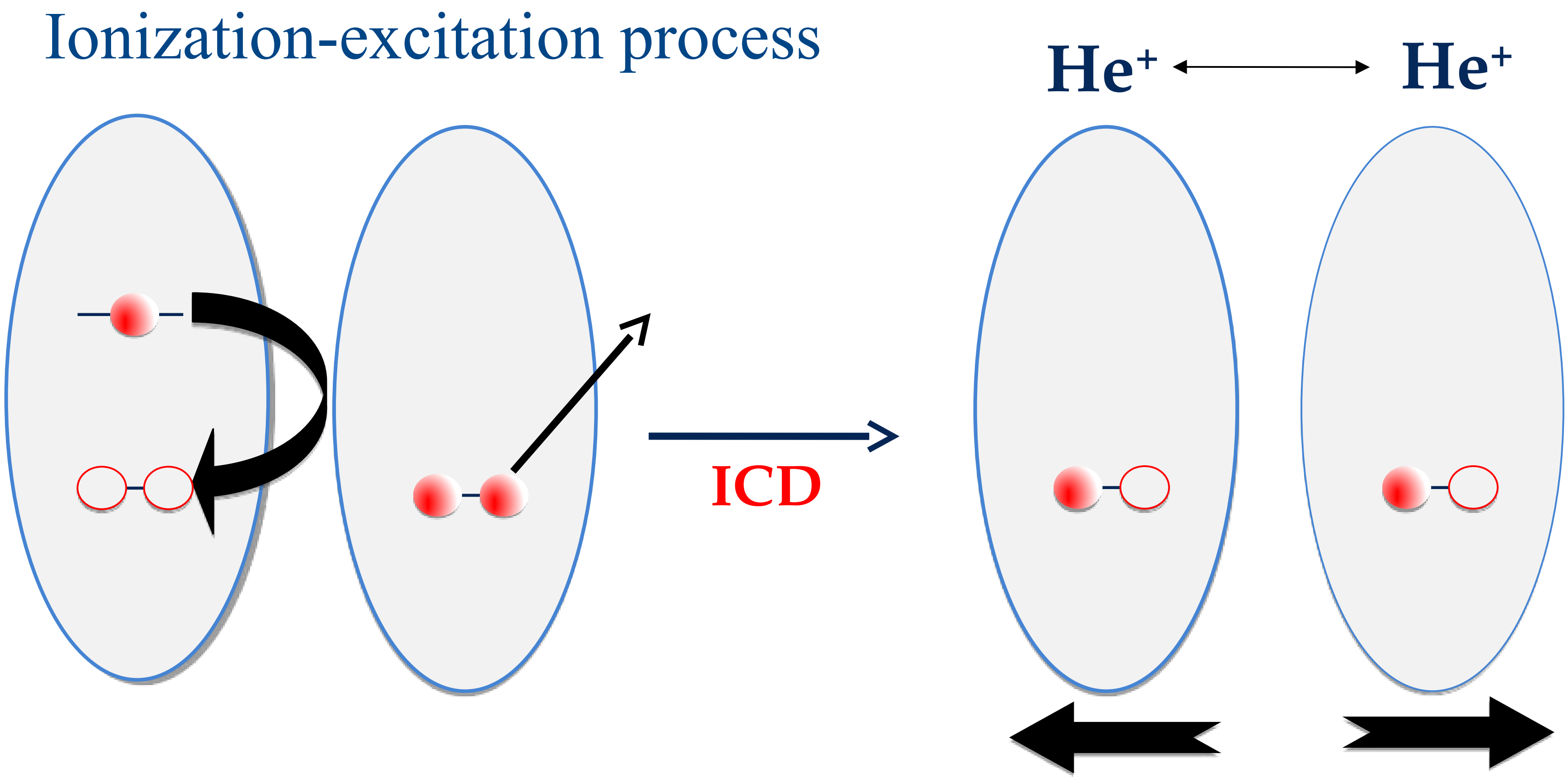
PHYSICAL REVIEW LETTERS **123**, 165301 (2019)

Featured in Physics

**Highly Charged Droplets of Superfluid Helium**

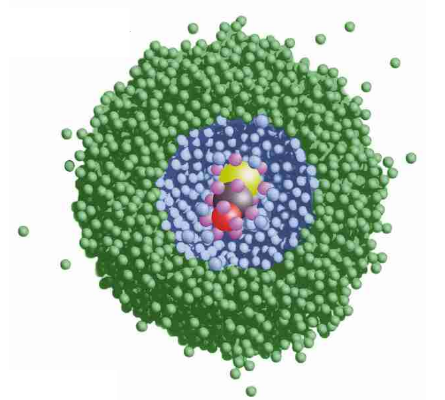
Felix Laimer,[1] Lorenz Kranabetter,[1] Lukas Tiefenthaler,[1] Simon Albertini,[1] Fabio Zappa,[1,2]
Andrew M. Ellis,[3] Michael Gatchell,[1,4,*] and Paul Scheier[1]

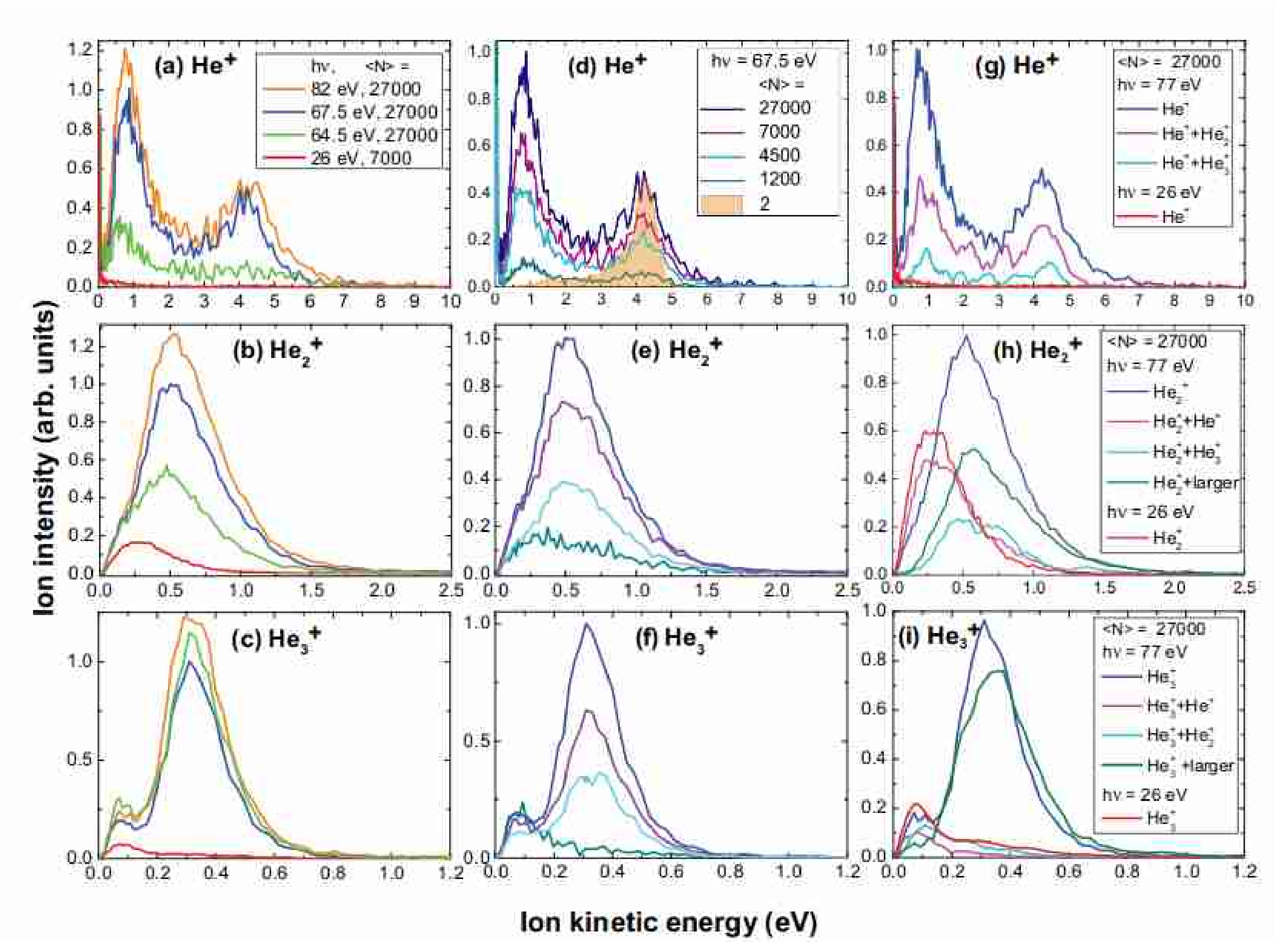
# ICD in helium clusters

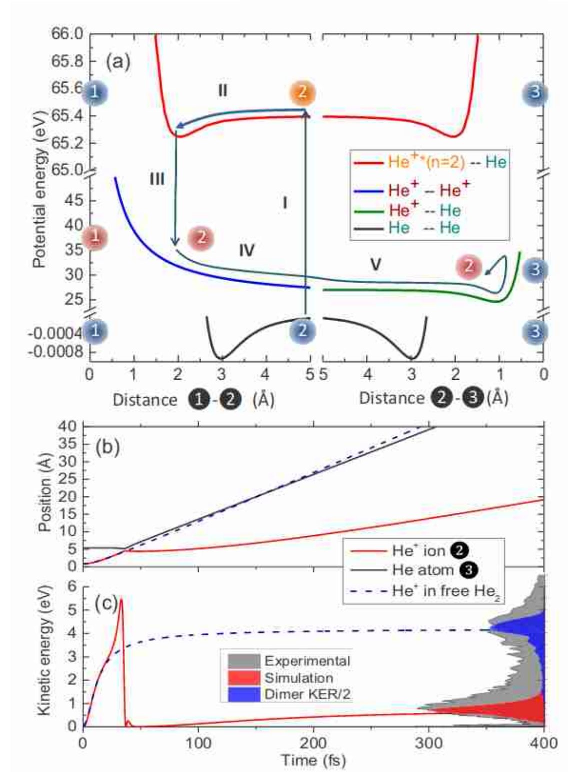



T. Havermeier, R. Dörner et al., Physical Review Letters **104**, 133401 (2010)
N. Sisourat, L.S. Cederbaum et al., Nature Physics **6**, 508 (2010)

# ICD in helium nanodroplets



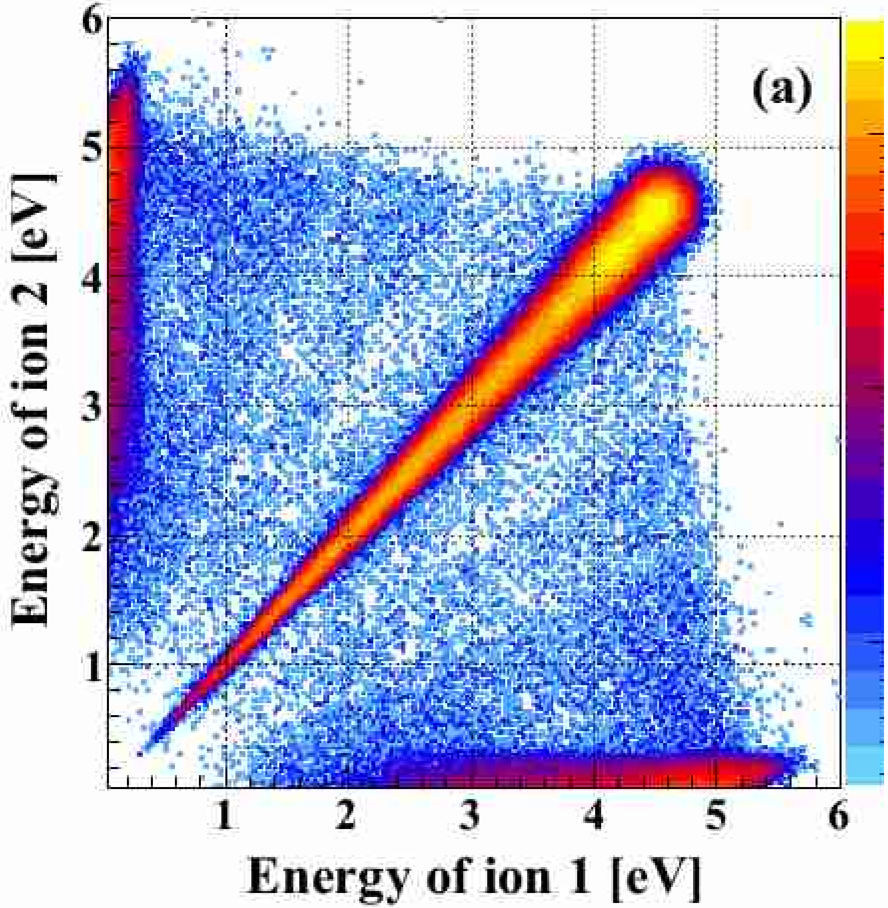



M. Shcherbinin, A. C. LaForge, V. Sharma, M. Devetta, R. Richter, R. Moshammer, T. Pfeifer, and M. Mudrich Phys. Rev. A **96**, 013407 (2017).

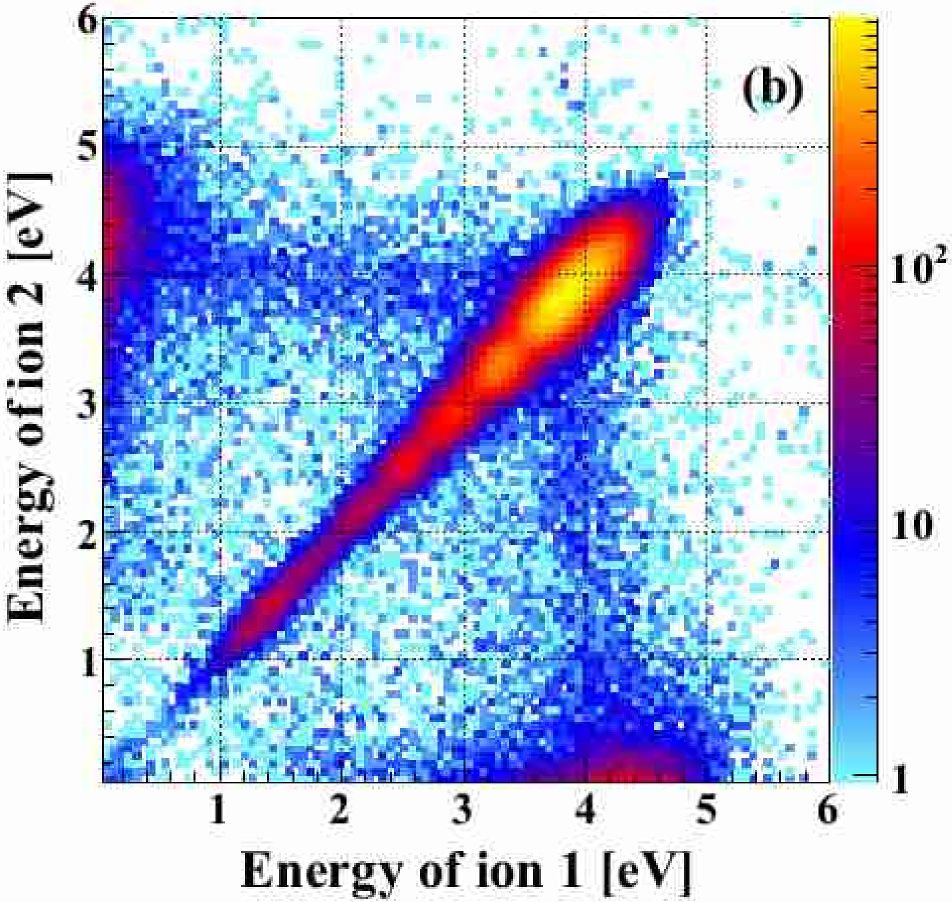
# $He_3$

Theory



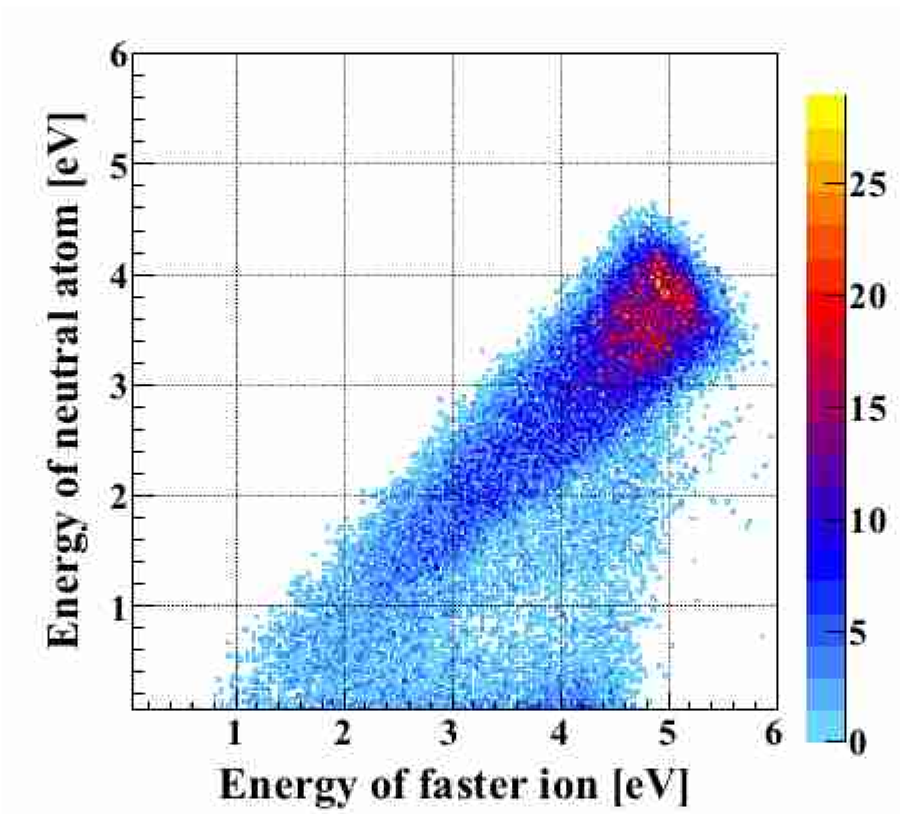

Expt.

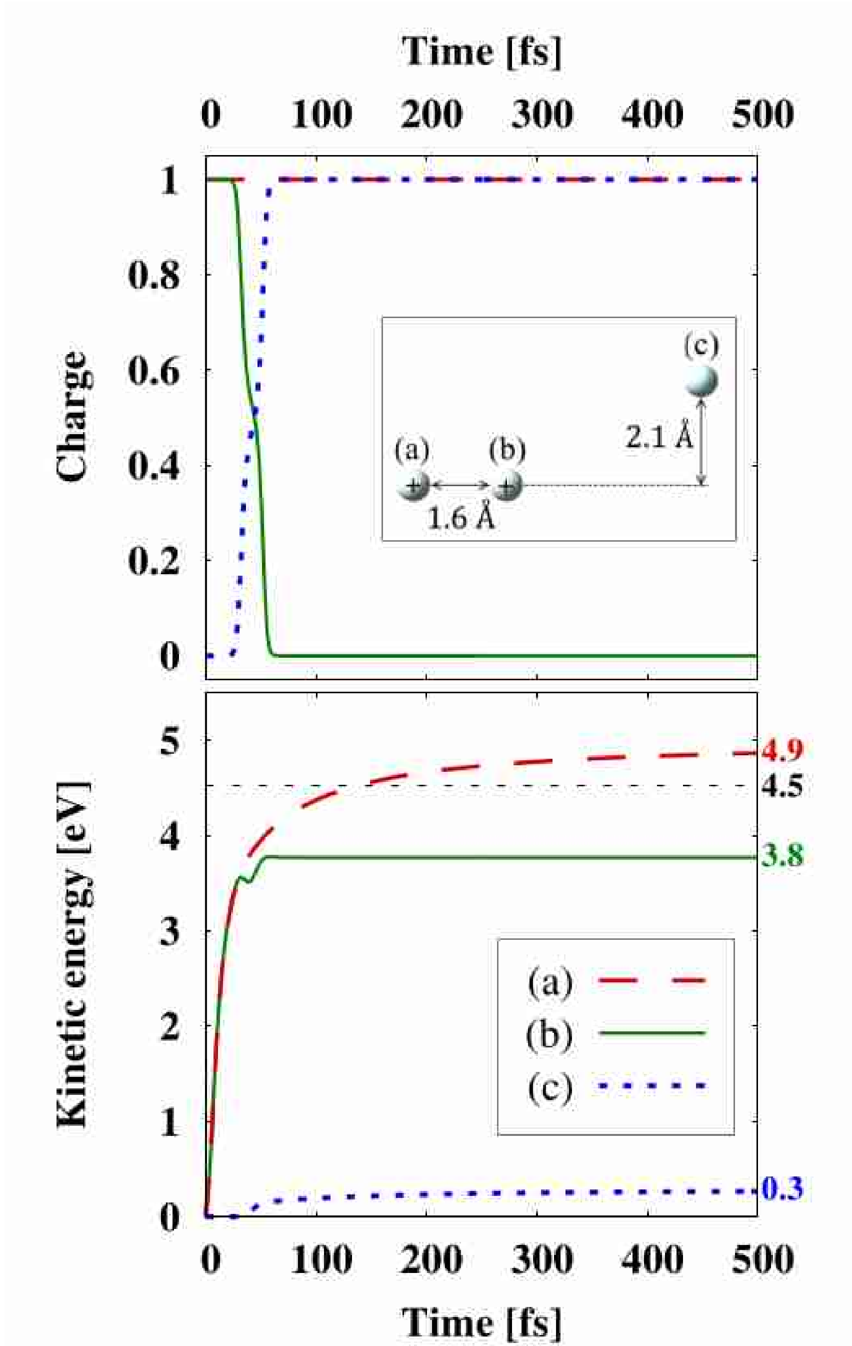
COLTRIMS (Frankfurt group)




Phys. Rev. A **98**, 050701(Rapid. Comm.) (2018)

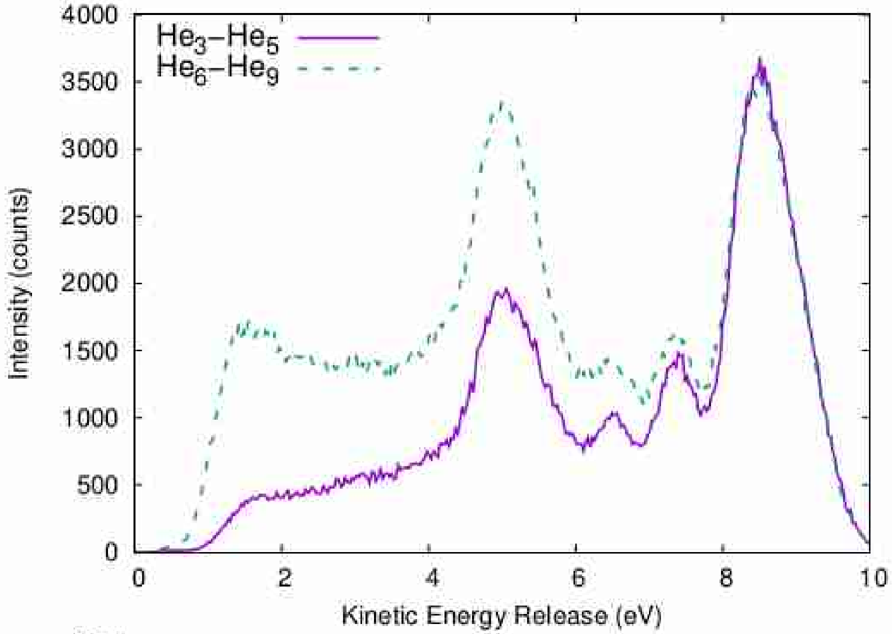
# $He_3$

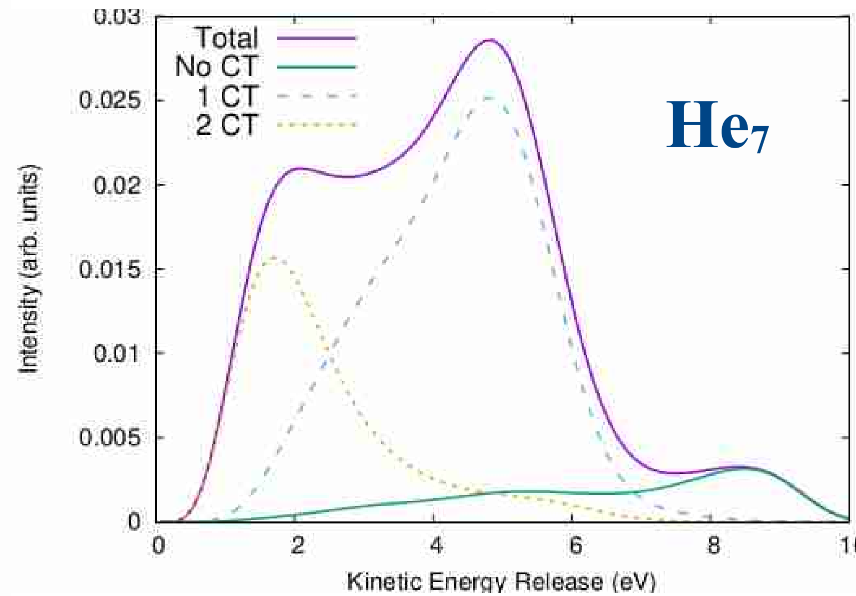


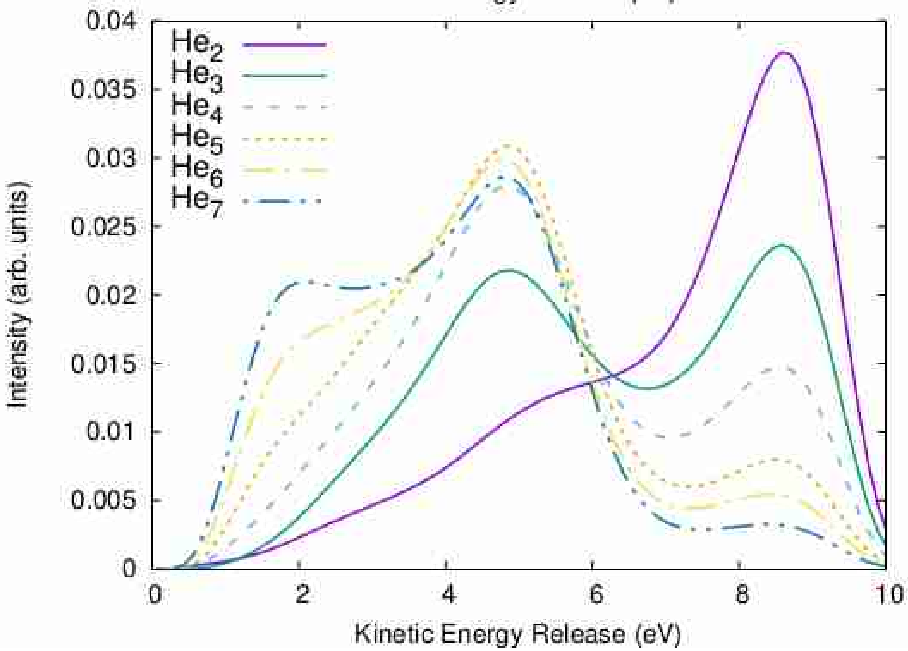




Phys. Rev. A **98**, 050701(Rapid. Comm.) (2018)

# $He_{2-7}$



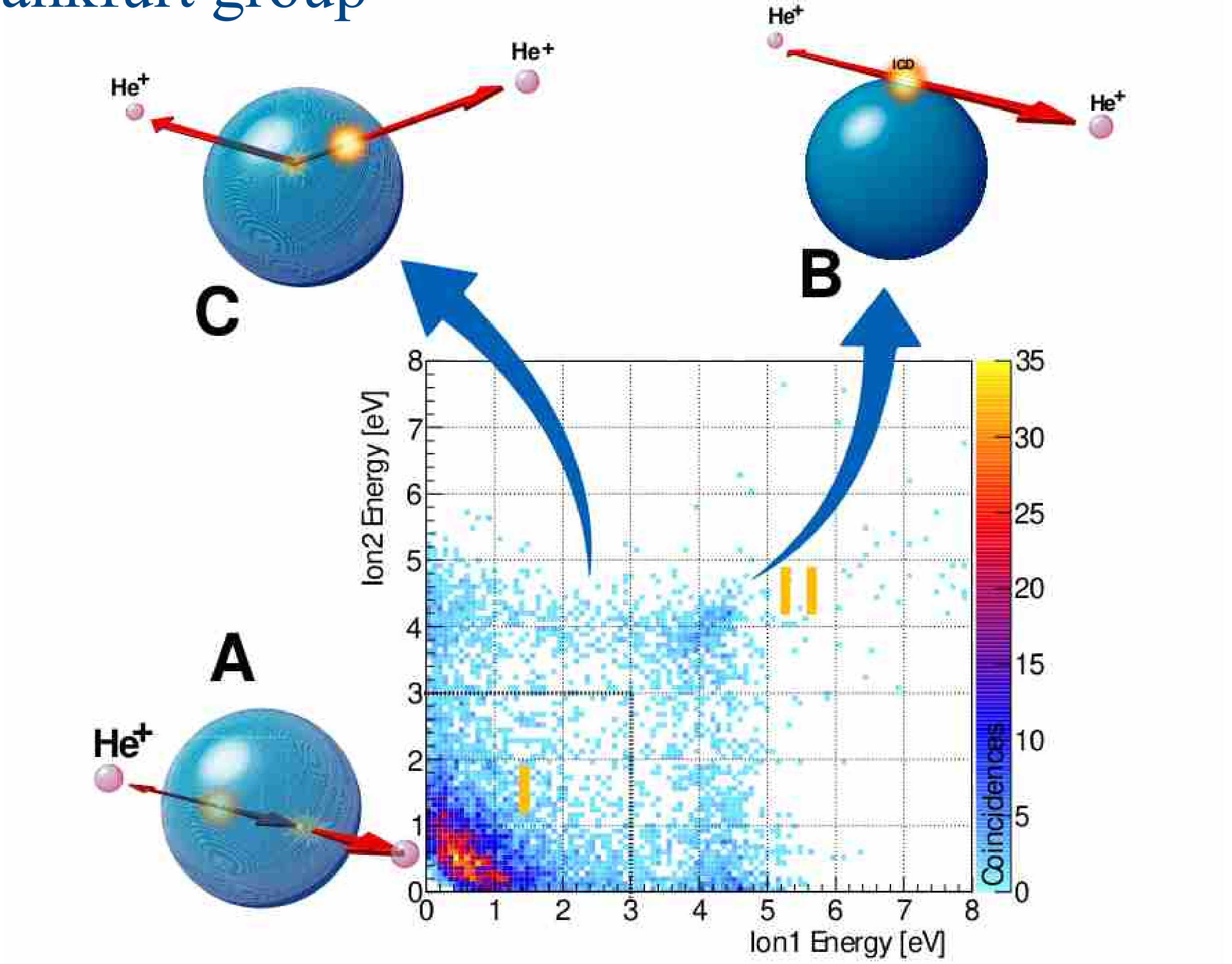




| $N$ | 2 | 3 | 4 | 5 | 6 | 7 |
|---|---|---|---|---|---|---|
| No charge transfer | 1.00 | 0.69 | 0.45 | 0.25 | 0.17 | 0.11 |
| One charge transfer | 0.00 | 0.31 | 0.51 | 0.63 | 0.64 | 0.62 |
| Two charge transfers | 0.00 | 0.01 | 0.04 | 0.12 | 0.19 | 0.27 |

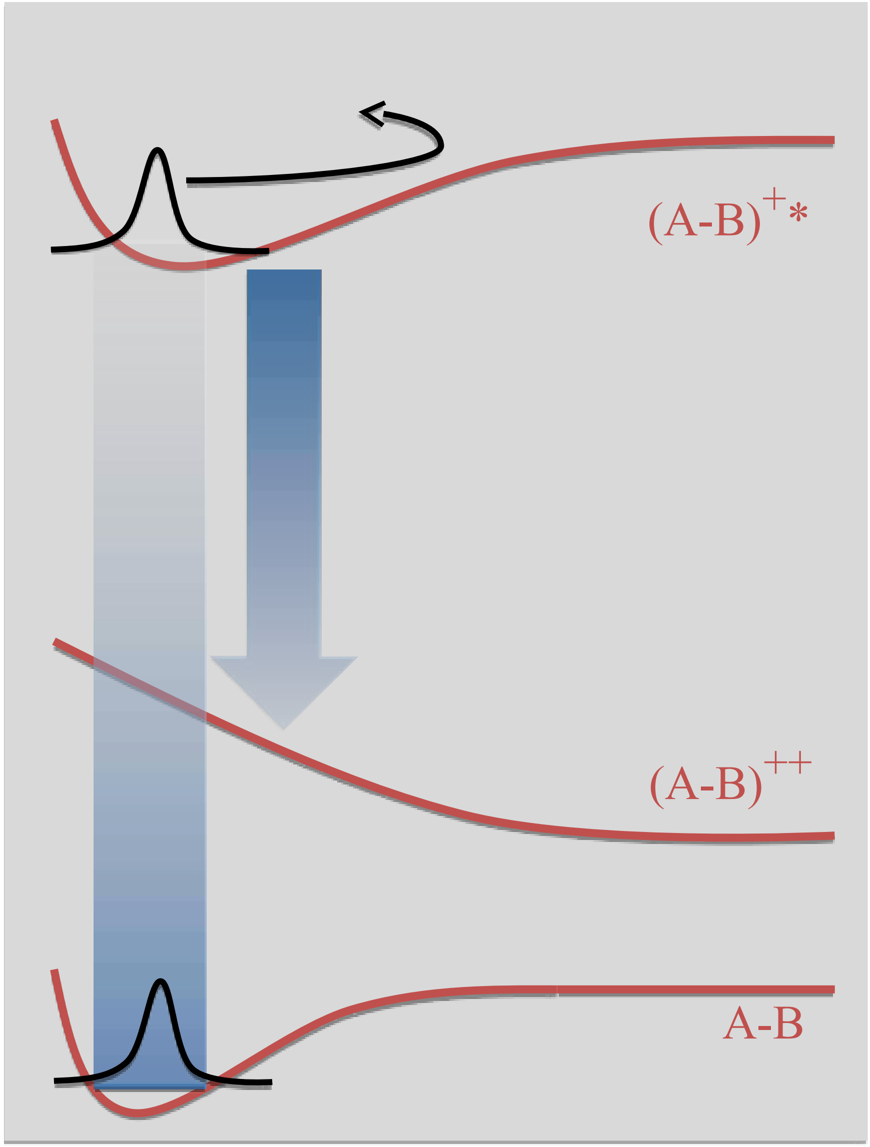
# Larger clusters
(~5000-30000atoms)

Results from the Frankfurt group




Phys. Rev. A **100**, 022707 (2019)

# How to describe ICD?

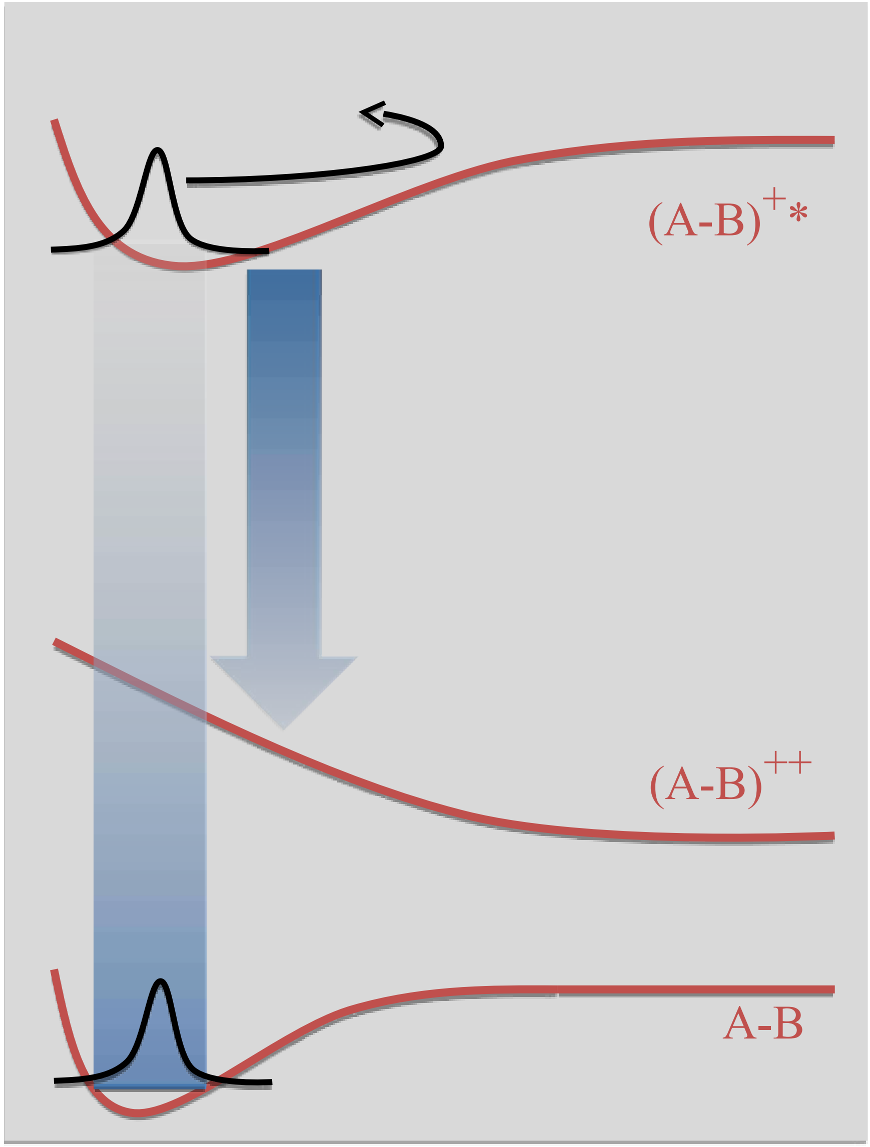



$$i\,|\dot{\phi}_d(t)\rangle = \left(H_d - i\frac{\Gamma_d}{2}\right)|\phi_d(t)\rangle$$

$$\sigma_{KER}(E_{KER}) = 2\pi\int_0^\infty |\langle E_f|W_{df}|\phi_d(t)\rangle|^2 dt$$

L. S. Cederbaum and F. Tarantelli. J. Chem. Phys. **98**, 9691 (1993).
Y.-C. Chiang et al. Phys. Rev. Lett. **107**, 173001 (2011).

# How to describe ICD?

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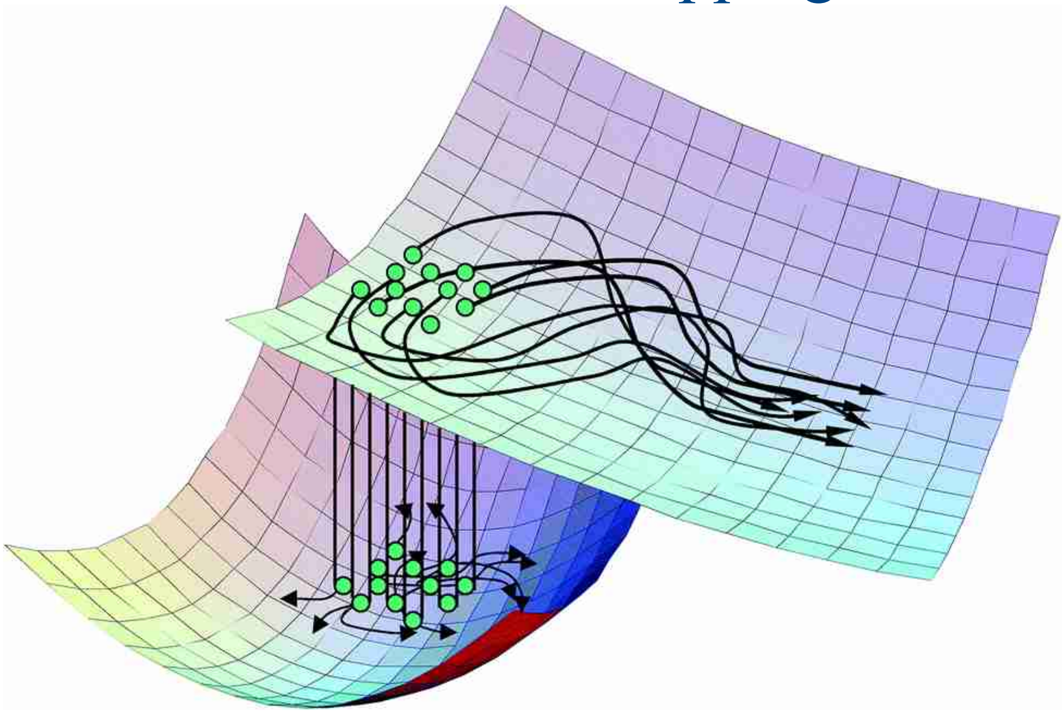
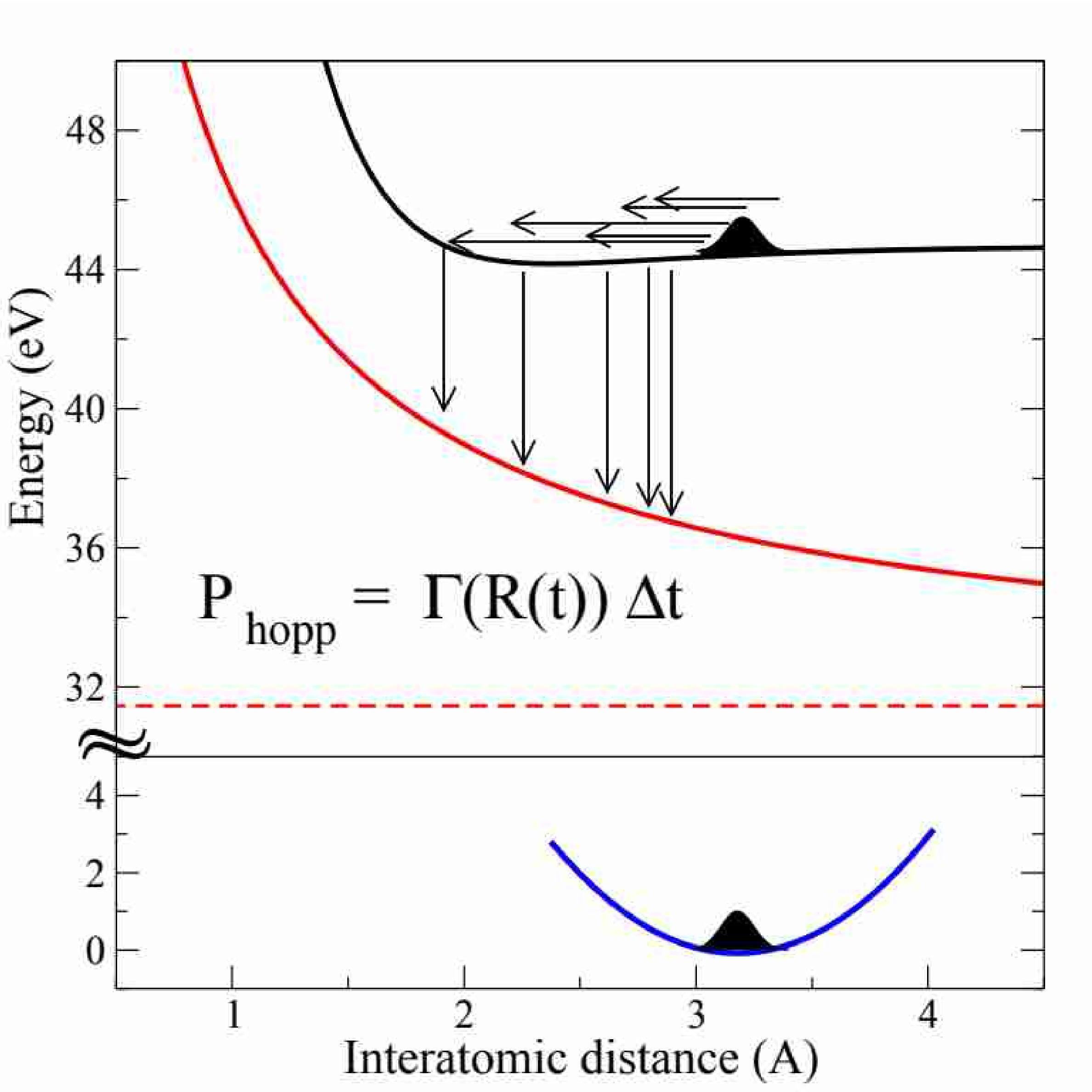
Surface hopping



N. Sisourat, J. Chem. Phys. **139,** 074111 (2013)
J.C. Tully. J. Chem. Phys. **93**, 141061 (1990)

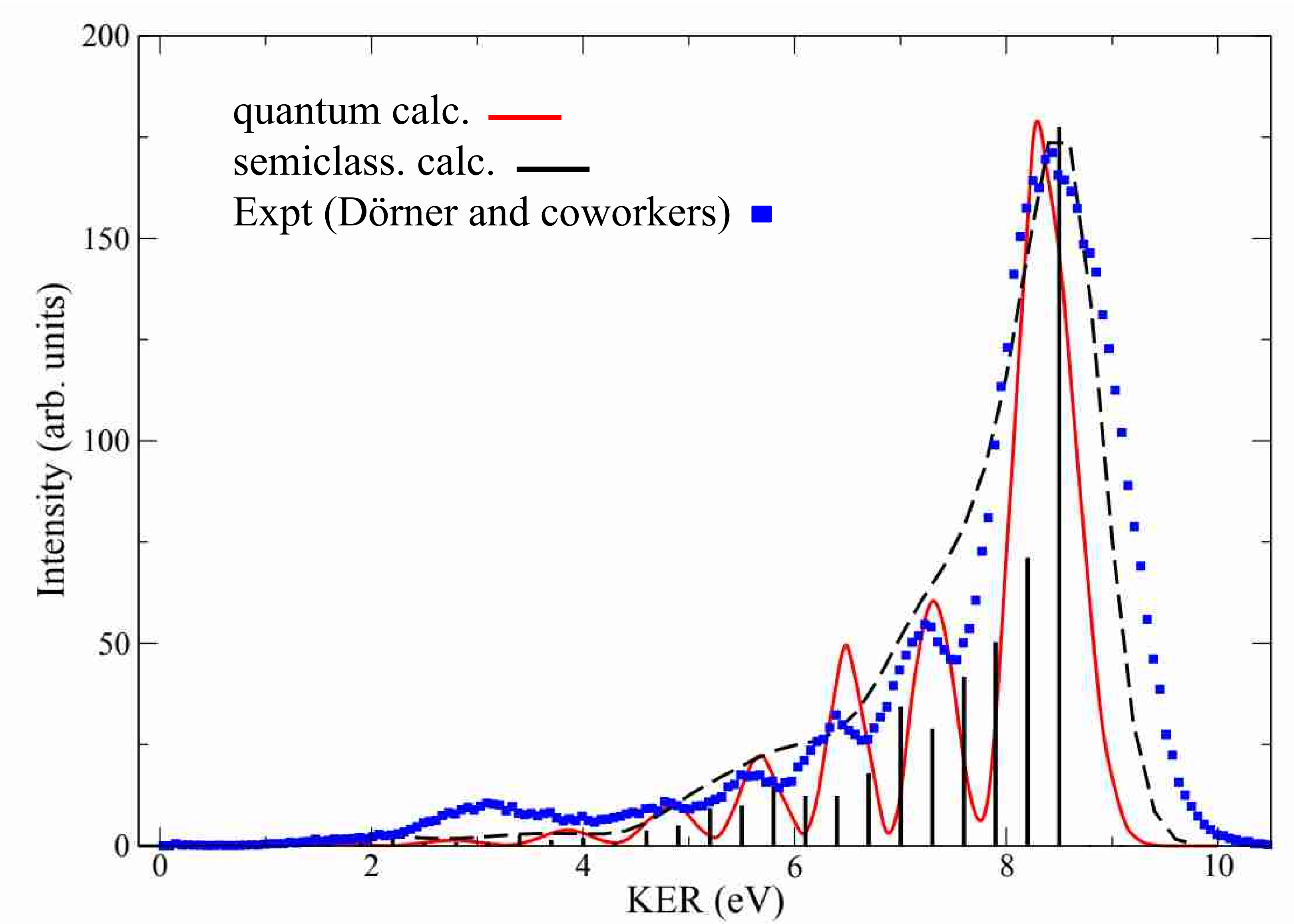
# Semiclassical method




Classical trajectories for nuclear Motion
+
« quantum » probabilities of « hopping »

N. Sisourat, J. Chem. Phys.,**139,** 074111 (2013)

# The helium dimer test




N. Sisourat, J. Chem. Phys.,**139,** 074111 (2013)

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Freiburg group
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Rob Bennett

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Tsveta Miteva
**Sévan Kazandjian**

ICD forschergruppe
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Petr Slavicek


Thank you for your attention

# Surface hopping *à la Tully*

Probability switch :

$$P_{df} = \frac{\dot{a}_f \Delta t}{a_d}$$

$a_f$ and $a_d$ = population of final and decaying state respectively

$$a_d = \langle \phi_d | \phi_d \rangle \quad + \quad i|\dot{\phi}_d(t)\rangle = (H_d - i\frac{\Gamma_d}{2})|\phi_d(t)\rangle$$

$$\Rightarrow \quad \dot{a}_d(t) = -\langle \phi_d | \Gamma_d | \phi_d \rangle\, a_d$$

Semiclassical approximation: $\langle \phi_d | \Gamma_d | \phi_d \rangle = \Gamma_d(R(t))$

$$a_d(t) = a_0 e^{-\int_0^t \Gamma_d(R(t'))t'}$$

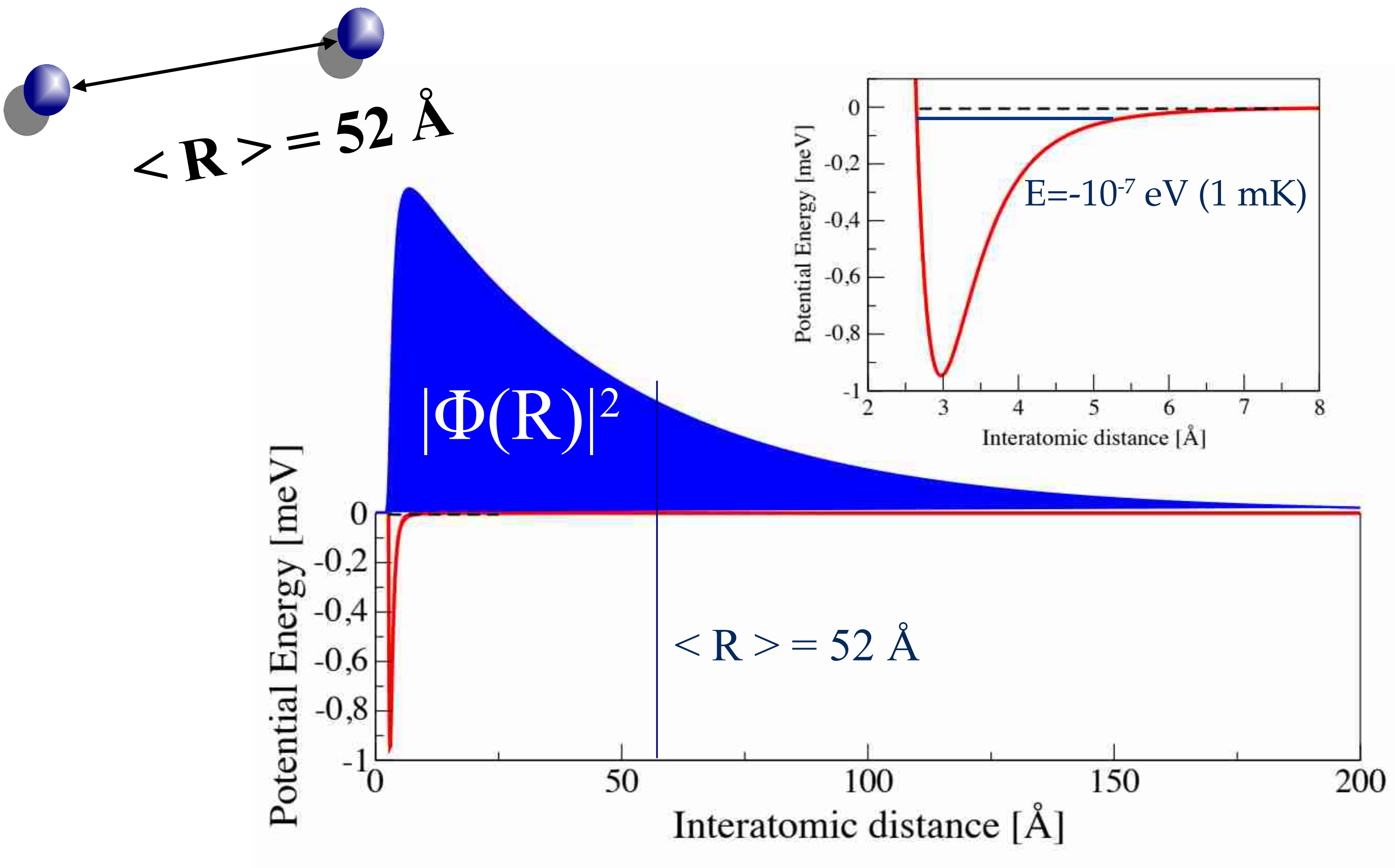
$$(a_d(t) + a_f(t) = \text{cte})$$

$$a_f(t) = a_0(1 - e^{-\int_0^t \Gamma_d(R(t'))t'})$$

$$P_{df}(t) = \Gamma_d(R(t))\Delta t$$

N. Sisourat, J. Chem. Phys.,**139,** 074111 (2013)
J.C. Tully. J. Chem. Phys., **93**, 141061 (1990).

# ICD in helium dimer



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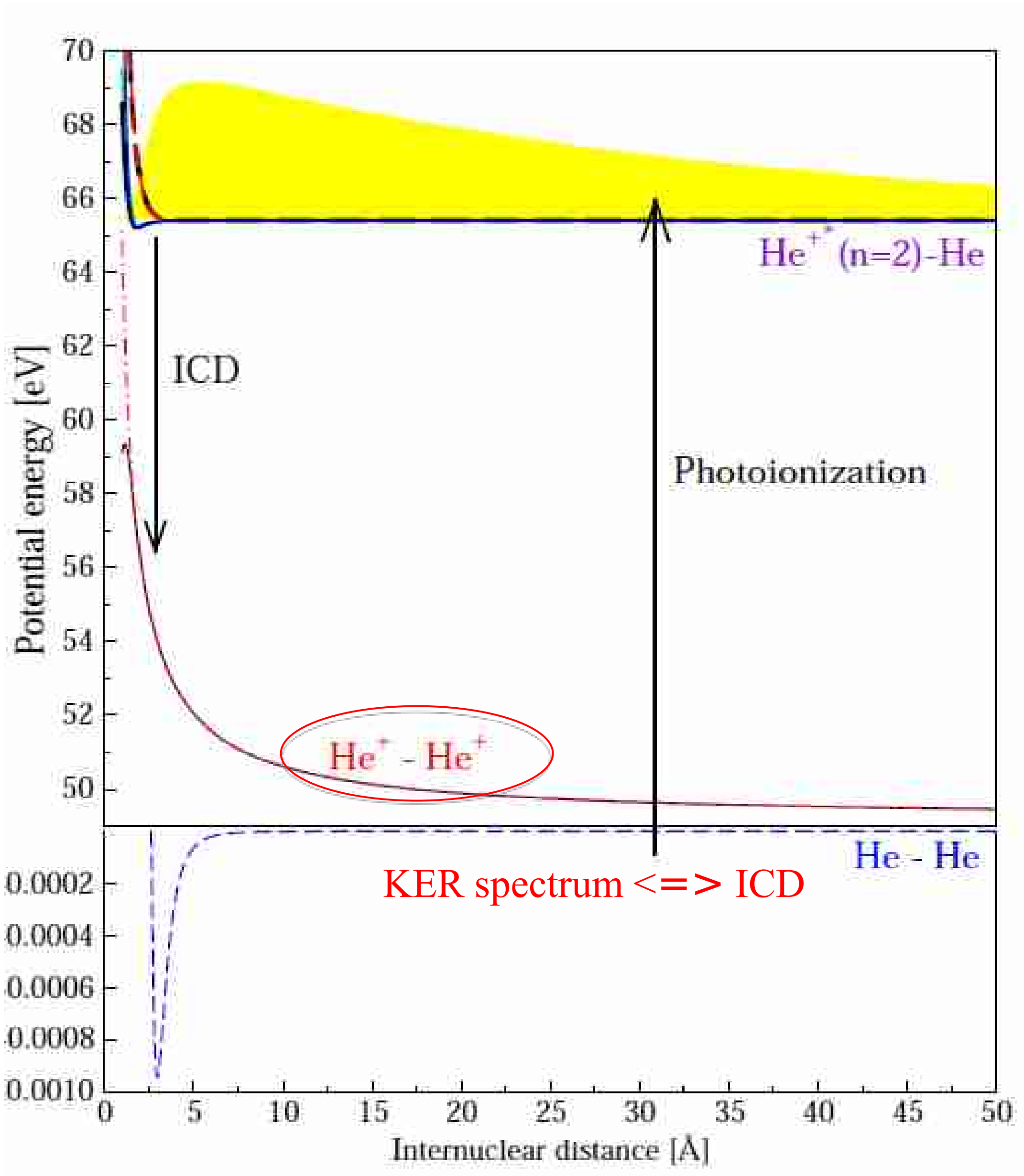
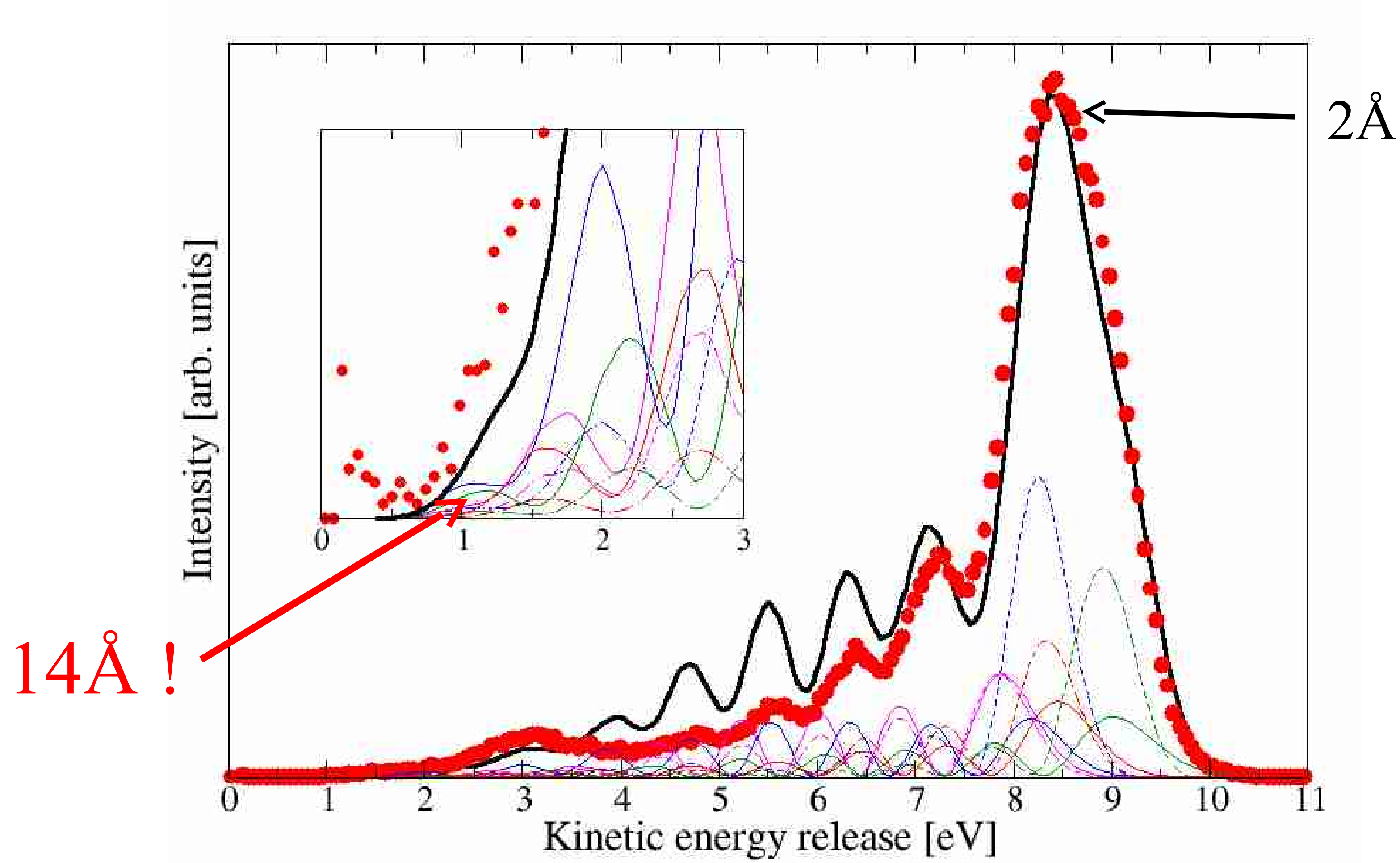


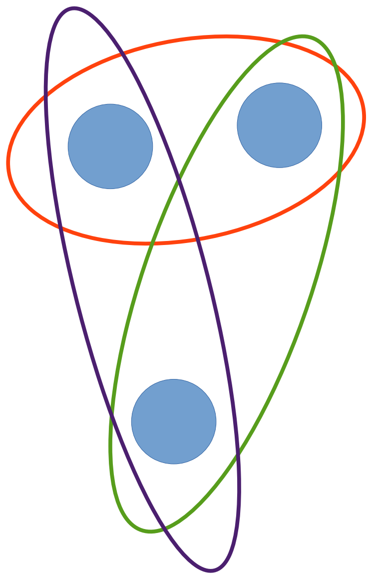
# ICD in helium dimer

## Total KER spectrum




T. Havermeier, R. Dörner et al., Physical Review Letters **104**, 133401 (2010)
N. Sisourat, L.S. Cederbaum et al., Nature Physics **6**, 508 (2010)

# Diatomics-in-molecules method



$$\hat{H}_e = \sum_{\alpha=1}^{N-1}\sum_{\beta>\alpha}^{N} \hat{H}^{\alpha\beta} - (N-2)\sum_{\alpha=1}^{N} \hat{H}^{\alpha}$$

$$|\Psi_m^{anti}\rangle = \hat{A}|\Psi_m\rangle = \hat{A}\hat{s}\prod_{n=1}^{N} |\chi_m^n\rangle$$

$$\hat{H}^{\alpha\beta} = \sum_i |\psi_i^{\alpha\beta}\rangle \epsilon_i^{\alpha\beta} \langle\psi_i^{\alpha\beta}|$$

$$\underline{\underline{H}}^{DIM} = \sum_\alpha\sum_\beta \underline{\underline{B}}^{\alpha\beta\dagger} \underline{\underline{\epsilon}}^{\alpha\beta} \underline{\underline{B}}^{\alpha\beta} - (N-2)\sum_\alpha \underline{\underline{e}}^{\alpha}$$

$$\underline{\underline{V}}_{d,f}^{DIM} = \underline{\underline{U}}\left[\sum_\alpha\sum_\beta \underline{\underline{B}}^{\alpha\beta\dagger} \underline{\underline{V}}_{d,f}^{\alpha\beta} \underline{\underline{C}}^{\alpha\beta} - (N-2)\sum_\alpha \underline{\underline{V}}_{d,f}^{\alpha}\right] \underline{\underline{W}}^{\dagger}$$

N. Sisourat, S. Kazandjian, A. Randimbiarisolo and P. Kolorenc, J. Chem. Phys. **144**, 084111 (2016).

# ICD widths in Helium trimer

| | $\Gamma_{DIM}$ ($10^{-5}$ a.u.) | $\Gamma_{\mathrm{Fano-ADC}}$ ($10^{-5}$a.u.) |
|---|---|---|
| $^2$A'($^1\Sigma_g^+/^2$P$_x$) | 1.21 | 1.46 |
| $^2$A'($^1\Sigma_g^+/^2$P$_z$) | 3.09 | 2.70 |
| $^2$A'($^2\Sigma_g^+/^1S$) | 2.89 | 2.67 |
| $^2$A'($^2\Sigma_u^+/^1S$) | 2.89 | 2.74 |
| $^2$A'($^2\Pi_g/^1S$) | 1.29 | 1.49 |
| $^2$A'($^2\Pi_u/^1S$) | 1.29 | 1.46 |
| $^2$A" ($^1\Sigma_g^+/^2$P$_y$) | 0.83 | 0.85 |
| $^2$A" ($^2\Pi_g/^1S$) | 0.80 | 0.83 |
| $^2$A" ($^2\Pi_u/^1S$) | 0.90 | 0.89 |

Total widths for equilateral geom. (R=4A)

N. Sisourat, S. Kazandjian, A. Randimbiarisolo and P. Kolorenc, J. Chem. Phys. **144**, 084111 (2016).