On the failure of theoretical descriptions of double ionization of helium by high energy electron impact

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Théorie, Modélisation, Simulation

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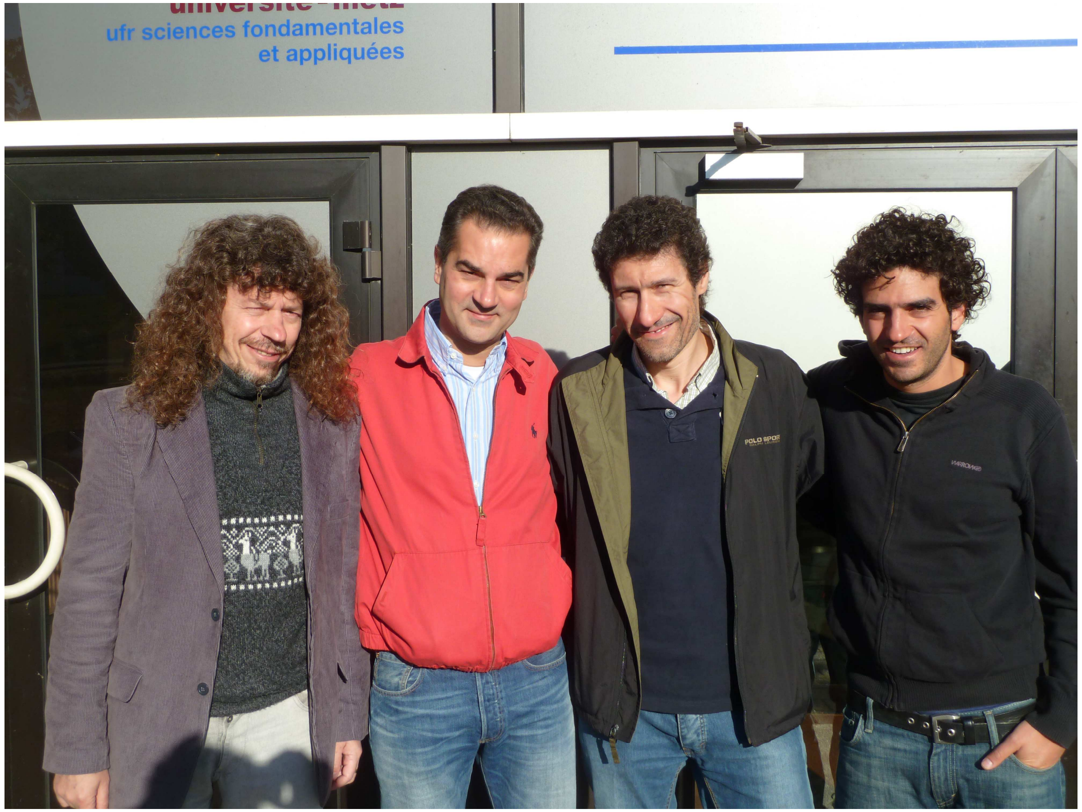

FINANCIAL SUPPORT:

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FRAMEWORK

- Atomic physics
- Coulomb three-body problem
- Configuration space

OUTLINE

- Ionization processes on He
  - What is working
  - What is not working

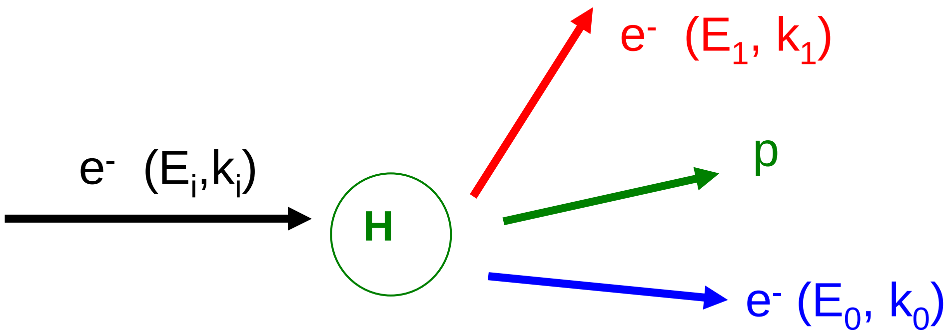
- Proposal of 2 three-body S-wave models to test numerical methods

  Method: Generalized Sturmian Functions

- Summary

SINGLE IONIZATION : (e,2e) on H

$e^- + H \longrightarrow p^+ + e^- + e^-$



Detection in coincidence:

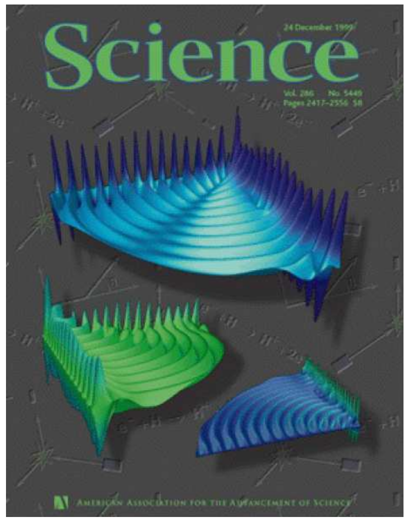
Kinematically complete

TDCS $$\frac{d^3\sigma}{d\Omega_1 d\Omega_0 dE_1}$$

- **Pure Coulomb 3-body problem in final channel**

- **Solved numerically at the end of century**
  [before that the Temkin-Poet model ($r_{12} \rightarrow r_>$)
  was used to test numerical methods !]

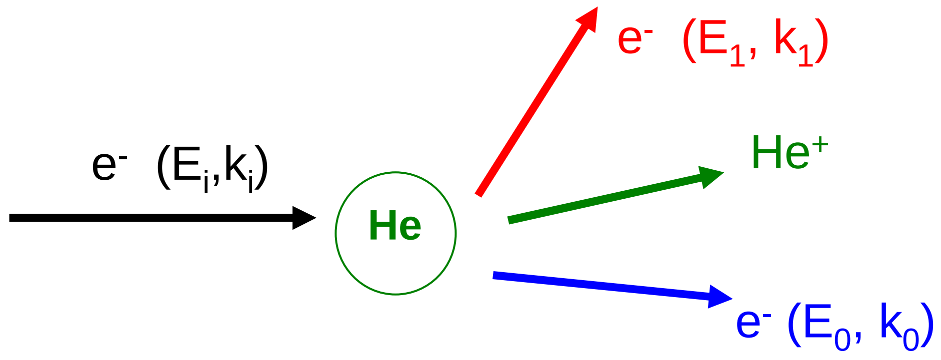
- **Agreement between theories and experiments**



From now: only He

SINGLE IONIZATION : (e,2e) on He

$e^- + He \longrightarrow He^+ + e^- + e^-$



Detection in coincidence:
Kinematically complete

TDCS $$\frac{d^3\sigma}{d\Omega_1 d\Omega_0 dE_1}$$

Pure 4-body problem
(6 interactions)
(3-body within FBA)

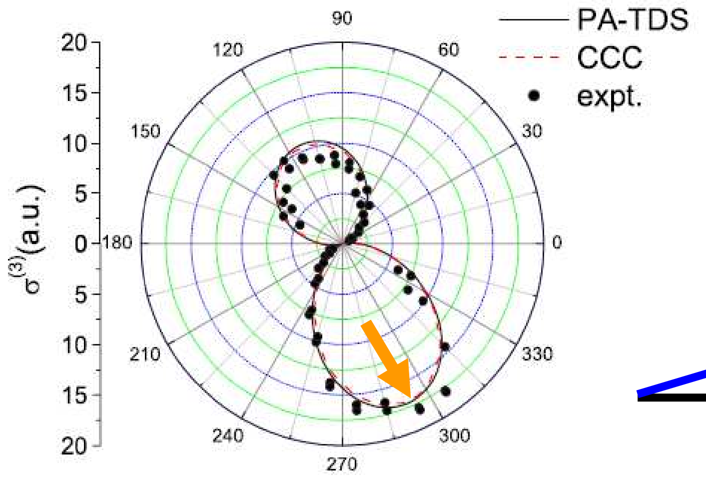
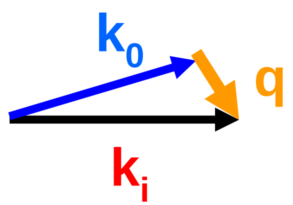


FIG. 4. (Color online) Single-ionization triple-differential cross section versus the ejection angle for the energy of impact electron, E_i=5600 eV; the scattering angle θ_s=0.45; and the energy of ejected electron, $E_{1,2}$=10 eV.

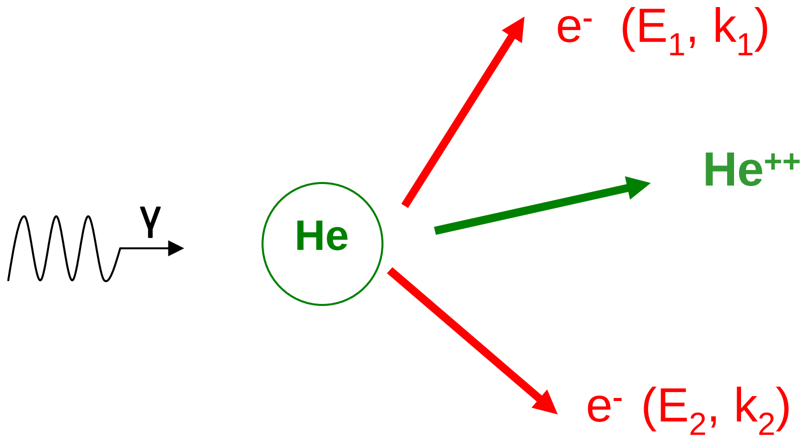


Agreement between 2 theories and with experiments

Serov et al (PRA, 2007)
Kheifets et al (JPB, 1999)

DOUBLE IONIZATION : (γ,2e)

$\gamma + He \longrightarrow He^{++} + e^- + e^-$

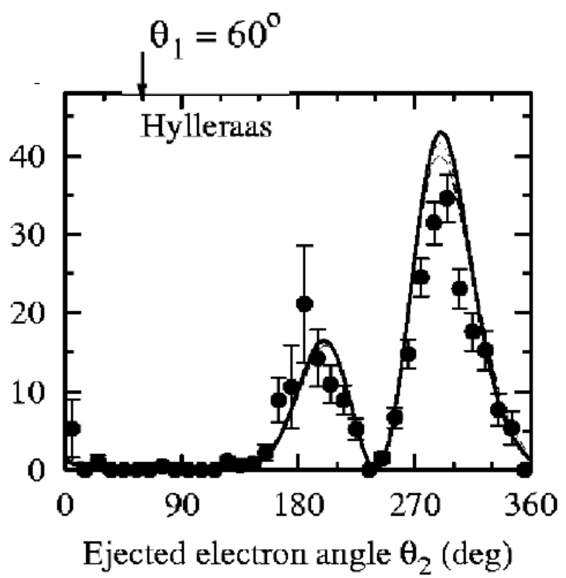


Pure 3-body problem
(3 interactions)

Detection in coincidence:
TDCS

$$\frac{d^3\sigma}{d\Omega_1 d\Omega_2 dE_2}$$

TDCS $E_1=E_2=10$ eV



CCC in three gauges
(Kheifets and Bray, PRA, 2004)

Agreement between 3 gauges
AND with other theories
AND with experiments ...
BUT tests only L=1

THEORY ⟷ EXPERIENCE

To calculate such cross sections one needs wave functions describing **three charged particles in Coulomb interaction $z_i z_j / r_{ij}$**

- Description of the bound state (target): OK
- Description of the double continuum: more difficult

Time Independent

Model

3C or 6C
with or without
effective charges

Numerical

- CCC
- ECS
- J-matrix
- HRM-SOW

Time Dependent

- TD Close Coupling
- Wave-packet evolution approach

Different *numerical* descriptions of the double continuum

agree reasonably well

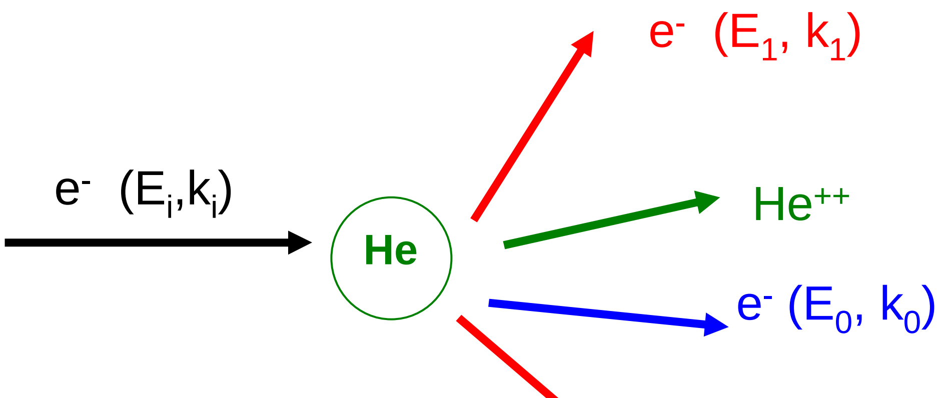
with each other AND with experiments

for (e,2e) and (γ,2e) processes

BUT … not for (e,3e) processes !

DOUBLE IONIZATION : (e,3e)

$e^- + He \longrightarrow He^{++} + e^- + e^- + e^-$



Pure 4-body problem
(6 interactions)

Detection in coincidence:
FDCS

$$\frac{d^5\sigma}{d\Omega_1 d\Omega_2 d\Omega_0 dE_1 dE_2}$$

Most stringent test for theory

Need - description of initial bound 3-body problem: OK
- description of continuum 4-body problem (or 3-body if within FBA)
(beautiful challenge for theoreticians)

Tests the double continuum in a complete manner

(e,3e) COPLANAR EXPERIMENTS on Helium

Absolute differential cross sections have been measured

- Measurements of Orsay Group

  (Lahmam-Bennani et al - PRA, 1999 and JPB,1999)

- Incident energy: E_i~5.6 keV

  Ejected energy: A) E_1=E_2=10 eV - q = 0.24 a.u.

  B) E_1=E_2=4 eV - q = 0.22 a.u.

  Dipolar regime: small momentum transfer $\mathbf{q}= |\mathbf{k}_i-\mathbf{k}_0|$

  (optical limit –> similar to (γ,2e))

FBA should be fine for these high incident energy AND small momentum transfers

(SECOND BORN EFFECTS are NEGLIGIBLE)

First Born Approximation (FBA)

$$\frac{d^5\sigma}{d\Omega_0 d\Omega_1 d\Omega_2 dE_1 dE_2} = (2\pi)^4 \frac{k_0 k_1 k_2}{k_i} \left| T_{fi}(\mathbf{k}_0, \mathbf{k}_1, \mathbf{k}_2) \right|^2$$

Interaction

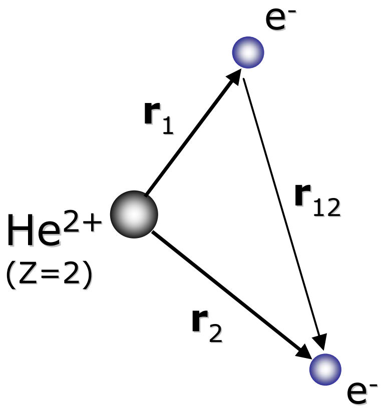
$$T_{fi} = \left\langle \frac{1}{(2\pi)^{3/2}} e^{i\mathbf{k}_0 \cdot \mathbf{r}_0} \Psi_f^-(\mathbf{r}_1, \mathbf{r}_2) \left| -\frac{Z}{r_0} + \frac{1}{r_{01}} + \frac{1}{r_{02}} \right| \frac{1}{(2\pi)^{3/2}} e^{i\mathbf{k}_i \cdot \mathbf{r}_0} \Psi_i(\mathbf{r}_1, \mathbf{r}_2) \right\rangle \quad \to \int 9D$$

Momentum transfer : $\mathbf{q} = \mathbf{k}_i - \mathbf{k}_0$

$$T_{fi} = \frac{1}{2\pi^2 q^2} \left\langle \Psi_f^-(\mathbf{r}_1, \mathbf{r}_2) \left| -Z + e^{i\mathbf{q}\cdot\mathbf{r}_1} + e^{i\mathbf{q}\cdot\mathbf{r}_2} \right| \Psi_i(\mathbf{r}_1, \mathbf{r}_2) \right\rangle \quad \to \int 6D$$

3-body CONTINUUM problem

(double continuum)



3-body BOUND problem
Ground state of He

No theoretical calculations reproduce all the experimental data !!

Ongoing debate for 14 years !

(Ancarani et al, J. Conf. Ser., 2010)

More than 15 papers with theoretical calculations

Orsay Measurements

E_i~5.6 keV

E_1=E_2= 4 eV

θ_0 =0.45°

16 angles θ_1

CCC calculations

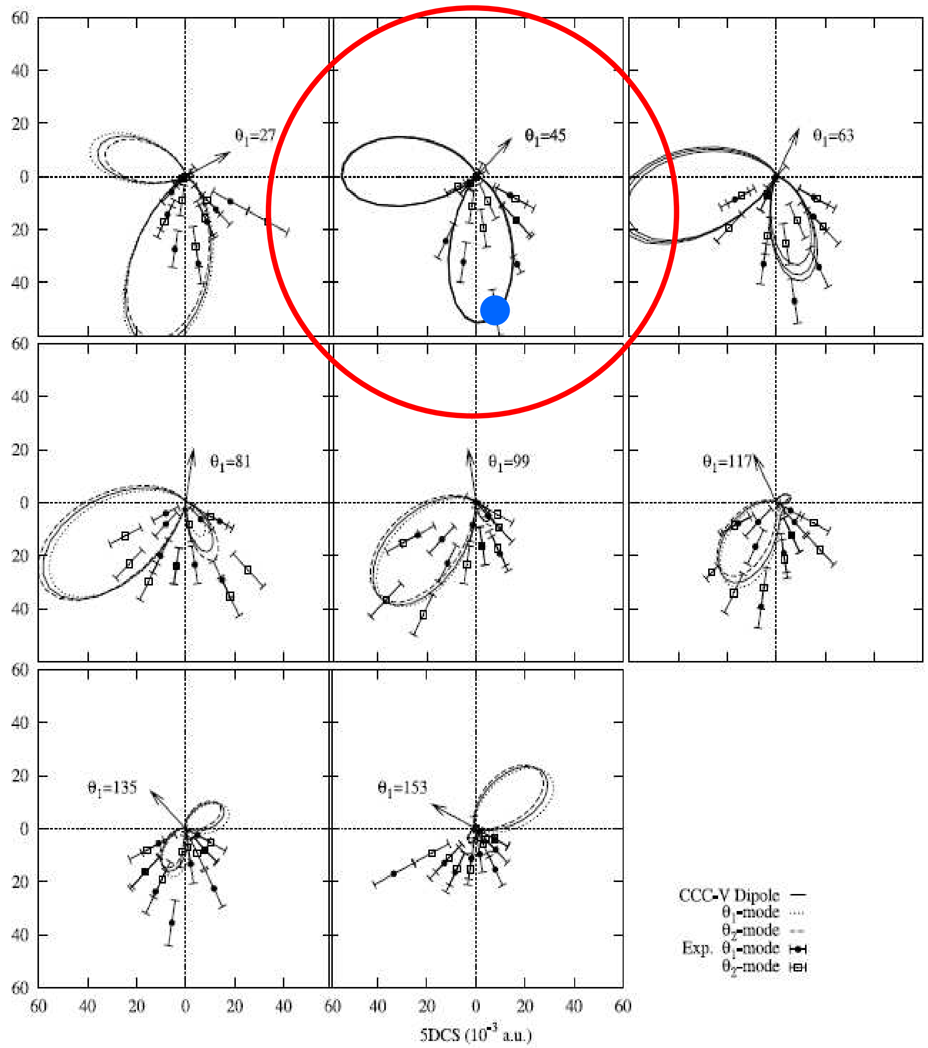
(with 20-term Hylleraas He w.f.)

Kheifets et al (JPB, 1999)

Shapes: not so good

AND factor 14 too small !

(rescaled at θ_1=45^0)



No agreement between calculations and experiments,

in particular with respect to the magnitude (but not only).

The measured (e,3e) cross sections are **absolute**.

Their validity can be questioned …

but theories do not agree with each other !!

PRESENTLY: numerical methods

which agree with each other for (e,2e) and (γ,2e)

do not agree for (e,3e) !!

- **IS SOMETHING GOING WRONG WITH NUMERICAL METHODS when describing (e,3e) processes?**
- **WHAT IS THE NATURE OF THE PROBLEM?**

- **WHAT CAN BE DONE TO RESOLVE THE ISSUE?**

WHAT CAN BE DONE TO RESOLVE THE ISSUE?

- **EXPERIMENTALLY:** need new absolute (e,3e) measurements
  - confirm those of 1999
  - other energy values ($E_1=E_2$ or $E_1 \neq E_2$)

- **THEORETICALLY:**
  - investigate again (properly) Second Born effects
  - check all convergence issues of *numerical* methods
  - test the double continuum in a simpler manner

↳ THIS IS WHAT WE DID : 2 models

METHOD: Generalized Sturmian Functions

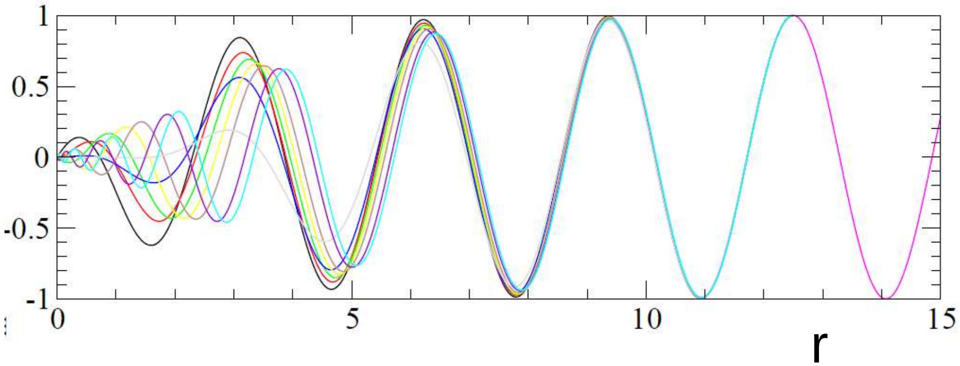
Generalized Sturmian Functions (two-body): $S_{n,l}(r)$

- solutions of a Sturm-Liouville differential equation
- they form a complete and discrete set → BASIS SET
- they have a unique and appropriate asymptotic behavior
  (with correct energy for continuum states)

Coulombic CONTINUUM states

$E=k^2/2$

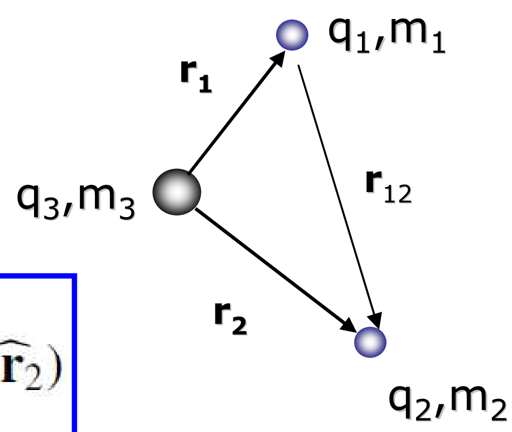
$$S^{\pm}_{n,l}(r) \rightarrow e^{\pm ikr \mp i\frac{Z_{as}}{k}\ln(2kr)}$$



- they concentrate the effort in the inner part where interaction takes place
- they transform the Schrödinger equation into a matricial problem

→ VERY EFFICIENT BASIS
(require smaller computational resources)

THREE-BODY CASE



Uncorrelated product of two-body GSF:

$$\Psi^{+}_{sc}(\mathbf{r}_1,\mathbf{r}_2) = \sum_{L\, l_1 l_2} \sum_{n_1} \sum_{n_2} a^{l_1 l_2 L}_{n_1 n_2}\, \mathcal{A}\, \frac{S^{+}_{n_1 l_1}(r_1)}{r_1} \frac{S^{+}_{n_2 l_2}(r_2)}{r_2} \mathcal{Y}^{L0}_{l_1 l_2}(\widehat{\mathbf{r}}_1,\widehat{\mathbf{r}}_2)$$

- **Similar advantages** (inner part, matricial problem, …)
- **Several successful applications**

➢ **BOUND** - atomic states (ground and excited), resonances

(e.g. BEST ground state energy with uncorrelated product)

- H_2^+ molecule
- confined systems

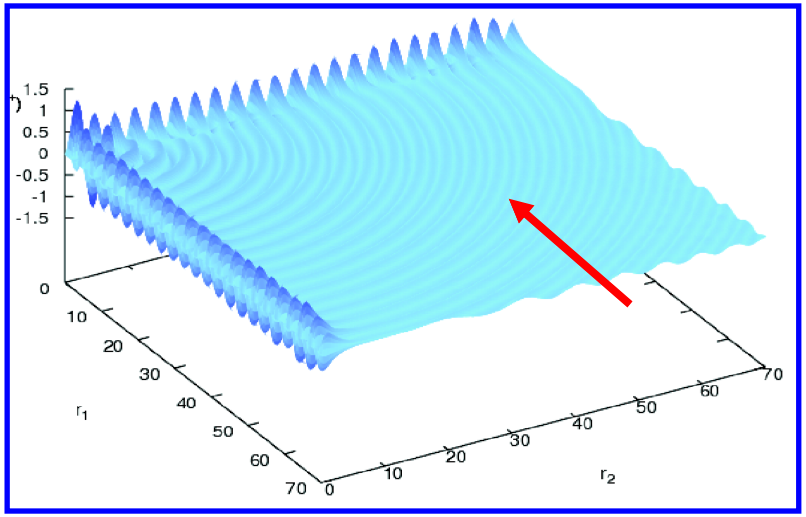
➢ **CONTINUUM**

- Single ionization of H by electron impact
- Single photoionization of H, He and CH_4 **(poster Granados)**
- Double photoionization of He

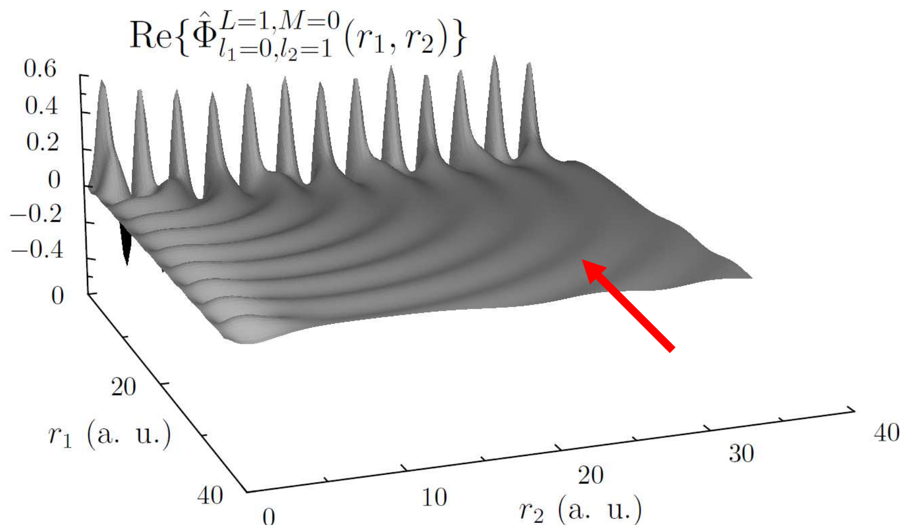
Mitnik et al, Comp. Phys. Comm., 2011 -- Gasaneo et al, Adv. Quantum Chem., 2013 and references therein

Scattering wave function : Re (Ψ_{sc}^{+})

(e,2e) on H



(γ,2e) on He



GSF: good agreement with other theoretical results

Peterkop-type asymptotic behavior
(all particles far from each other : Ω_0)

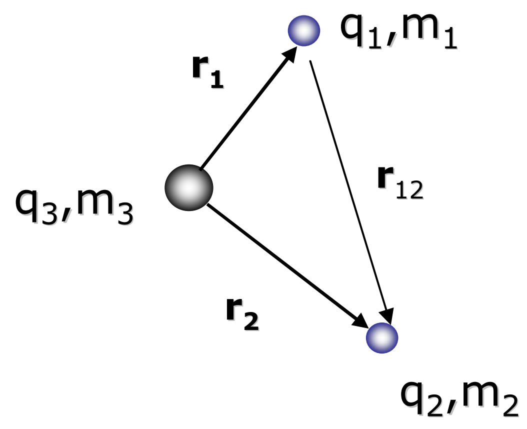
$$\Psi_{\mathrm{sc}}^{+}(\rho, \omega_5) = A(\omega_5)\rho^{-5/2} e^{iK\rho + i\frac{C(\omega_5)}{K}\ln(2K\rho)}$$

It is notoriously VERY difficult
to enforce Coulomb three-body asymptotic conditions

HYPERSPHERICAL COORDINATES: (ρ,ω_5)

$$\rho = \sqrt{r_1^2 + r_2^2} \qquad \omega_5 = \{\alpha, \hat{\mathbf{r}}_1, \hat{\mathbf{r}}_2\}$$

$$\alpha = \arctan(r_2/r_1)$$



$$V(r_1, r_2, r_{12}) = \frac{q_1 q_3}{r_1} + \frac{q_2 q_3}{r_2} + \frac{q_1 q_2}{r_{12}} \;\rightarrow\; V(\rho, \omega_5) = \frac{C(\omega_5)}{\rho}$$

Peterkop-type asymptotic behavior

$$\Psi_{\rm sc}^{+}(\rho, \omega_5) = A(\omega_5)\rho^{-5/2} e^{iK\rho + i\frac{C(\omega_5)}{K}\ln(2K\rho)}$$

Ionization amplitude

Angle-dependent coefficient of the logarithmic phase

Hyper-momentum $K = \sqrt{k_1^2 + k_2^2}$; $E = \dfrac{K^2}{2\mu}$

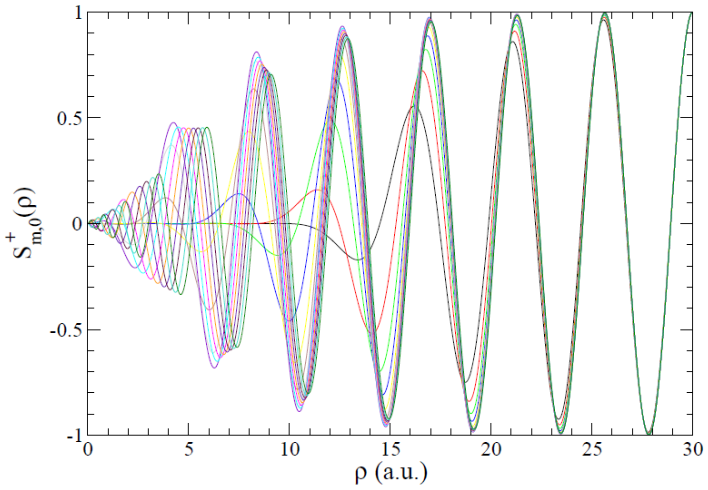
Hyperspherical coordinates (ρ,ω_5) are better suited (at least for the Ω_0 region) !!

→ **GENERALIZED STURMIAN FUNCTIONS (HGSF) IN HYPERSHERICAL COORDINATES** ⟶ $\left\{ \begin{array}{l} S^{+}_{n.m}(\rho) \\ \Omega_n(\omega_5) \end{array} \right.$

(Gasaneo et al, J.Phys.Chem. A, 2009, Gasaneo and Ancarani, J. Phys. A, 2012)

S-wave:

$$\Psi^{+}_{sc}(\rho,\alpha) = \frac{1}{\rho^{\frac{5}{2}}} \sum_m \sum_n a_{mn}\; S^{+}_{mn}(\rho)\; \Omega_n(\alpha)$$



Application to ionization processes

Solve $(H-E)\Psi = 0$

Set $\Psi = \Psi_0 + \Psi^{+}_{sc}$

Initial state
$(H+W-E)\Psi_0=0$

Scattering function

Driven Equation

$(H-E)\,\Psi^{+}_{sc} = -W\,\Psi_0$

Driven Equation in Hyperspherical coordinates (ρ,ω_5)

$$[T + V(\rho,\omega_5) - E]\Psi_{sc}(\rho,\omega_5) = -W(\rho,\omega_5)\Psi_0(\rho,\omega_5)$$

S-wave Models

Temkin-Poet : **Dielectronic interaction** r_{12} → $r_> = \mathrm{Max}[r_1, r_2]$

$$V(\rho, \omega_5) = \frac{C(\omega_5)}{\rho} \rightarrow \frac{C(\alpha)}{\rho}$$

Since this is OK for (e,2e) with several *numerical* methods
→ something else ?

Multiply the (benchmark) tests

→ two S-wave model problems

1) Three-body break-up model problem

2) Double ionization of He by high energy electron impact

1) Three-body break-up model problem

(Ancarani, Gasaneo and Mitnik, EPJD, 2012; Mitnik, Ancarani, Gasaneo, JPB, 2012)

Full three-body >> Temkin-Poet >> Present Model

$$V(\rho,\omega_5) = \frac{C(\omega_5)}{\rho}$$

$$\frac{C(\alpha)}{\rho}$$

$$\frac{q_1 q_2}{\rho}$$

$$\left[T - \frac{Z}{\rho} - E\right]\Psi(\rho,\alpha) = W(\rho,\alpha)\Psi_0(\rho,\alpha)$$

Bound-free initial state

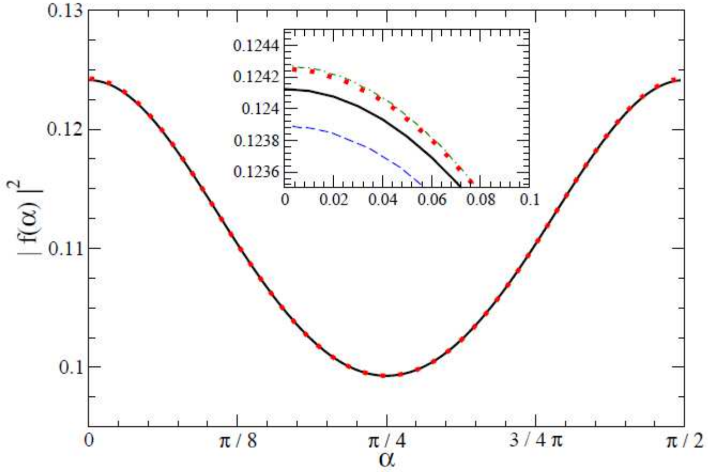
Same difficulties as real problem:

1) **Non separability**
2) **Coulomb potential**
3) **(r_1,r_2) coupling is different**

Three-body break-up model problem
has an **ANALYTICAL SOLUTION**

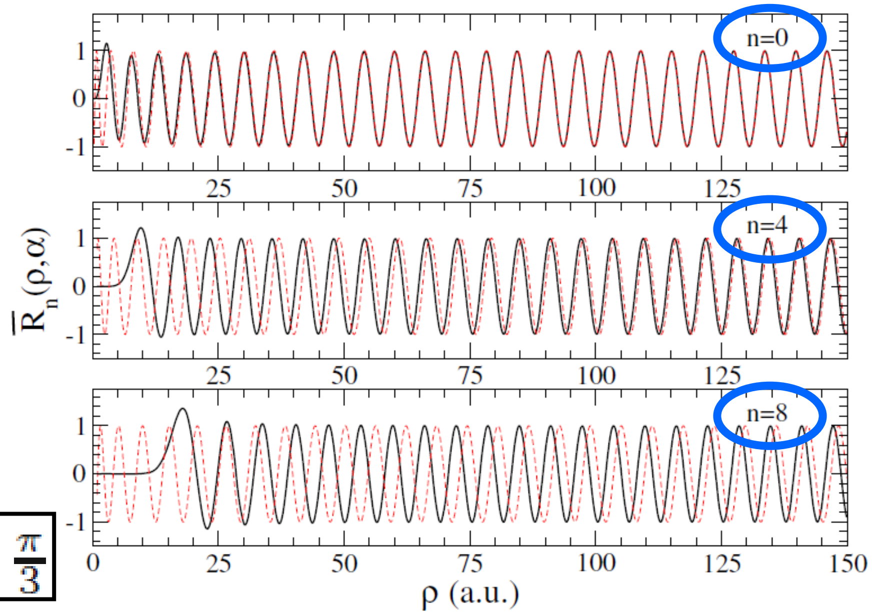
→ **analytical scattering amplitude f(α)**

→ **provides a solid three-body benchmark for testing numerical methodologies**



$K = 1$ a.u.
$E = 0.5$ a.u.

- **Excellent analytical-numerical agreement**
- **Hyperspherical GSF: good tool**
- **Very fast convergence (few n terms only)**
- **Also lower energy (usually convergence issues!)**

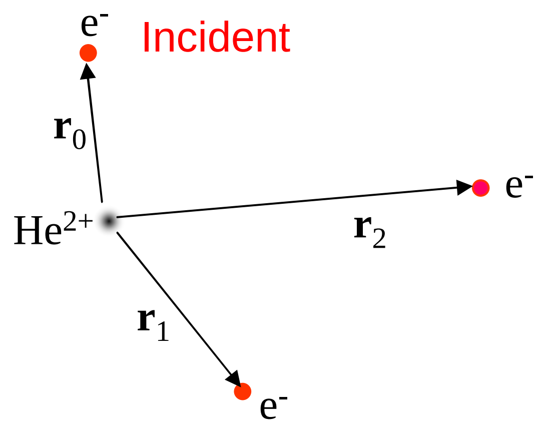


$\alpha = \frac{\pi}{3}$

To extract the transition amplitude
→**need to go very far !!**
(long range)

2) Double ionization of He by high energy electron impact

(**Gasaneo et al, PRA, 2013)**



- Four body problem
- Fast incident/scattered electron
  - → 3-body Driven Equation (equivalent to FBA)
- **S-wave model for the driven equation:** $r_{12} \rightarrow r_> = \mathrm{Max}[r_1, r_2]$

(e,3e) measurements on He of Orsay Group Incident energy: E_i~5.6 keV

Ejected energy: A) $E_1=E_2=10$ eV
B) $E_1=E_2=4$ eV

No other model at the energy of the (e,3e) experiments (5.6 keV – 10 eV, 10 eV)

Other S-wave models for ionization processes on He have been published, but focussed on lower incident energies.

We have solved this driven equation with:

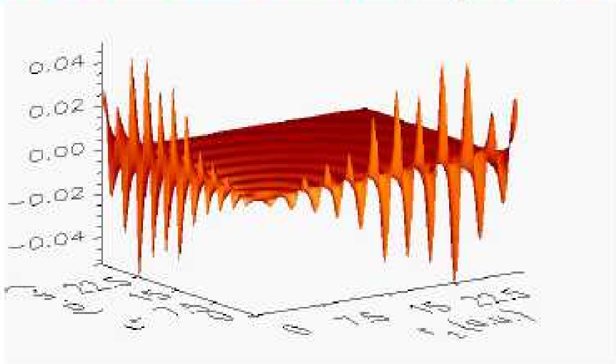
GSF

$$\Psi^+_{sc}(\mathbf{r}_1, \mathbf{r}_2) = \sum_{L\, l_1 l_2} \sum_{n_1} \sum_{n_2} a^{l_1 l_2 L}_{n_1 n_2} \mathcal{A} \frac{S^+_{n_1 l_1}(r_1)}{r_1} \frac{S^+_{n_2 l_2}(r_2)}{r_2} \mathcal{Y}^{L0}_{l_1 l_2}(\widehat{\mathbf{r}}_1, \widehat{\mathbf{r}}_2)$$

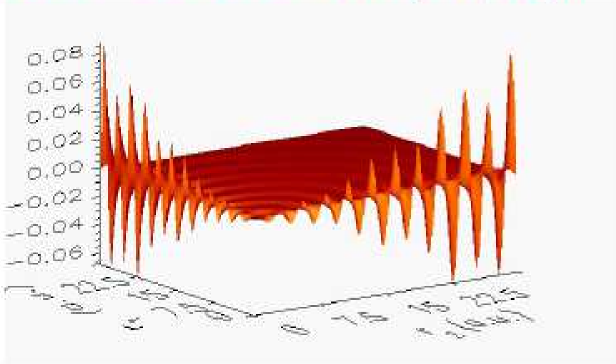
GHSF

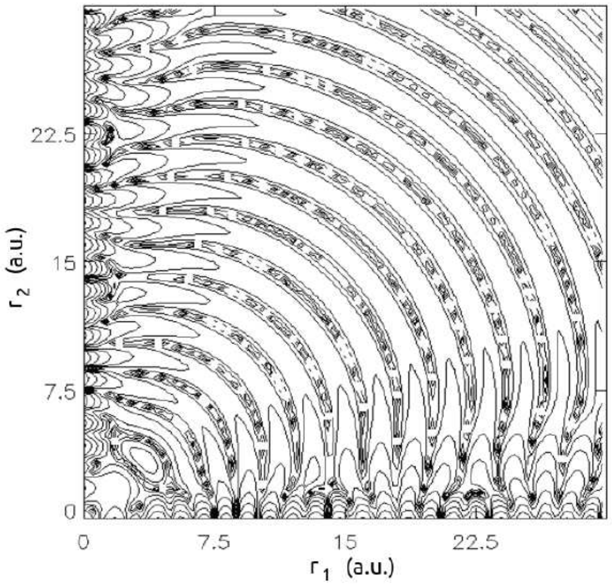
$$\Psi^+_{sc}(\rho, \alpha) = \frac{1}{\rho^{\frac{5}{2}}} \sum_m \sum_n a_{mn}\; S^+_{mn}(\rho)\; \Omega_n(\alpha)$$

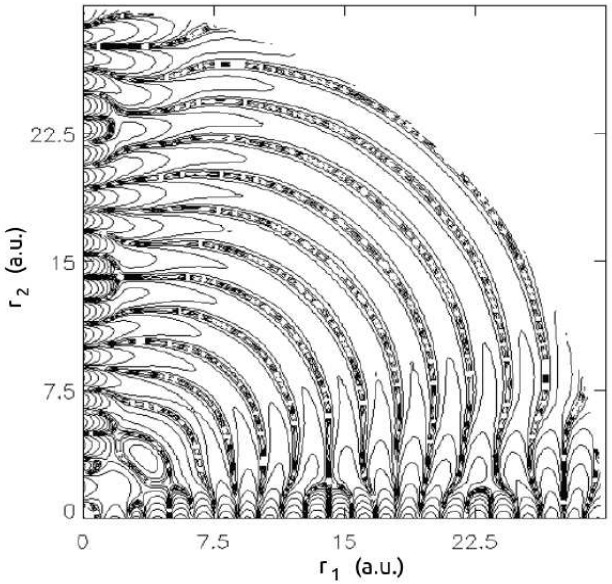
HGSF Calculation (ρ, α):



GSF Calculation (r_1, r_2):







+new benchmark SDCS data

TWO COMPLETELY INDEPENDENT numerical methods (GSF and GHSF) are in VERY GOOD AGREEMENT in describing the double continuum !!

Can other *ab initio* methods reproduce this simplified problem?

SUMMARY

- **PROBLEM: double ionization of He by high energy electron impact**
  (No agreement between theory and experiment AND between theories)
  THEORETICAL / EXPERIMENTAL REASONS??

- **METHOD: appropriate boundary conditions can be imposed to the basis elements**
  - spherical GSF are able to generate the correct outgoing 3-body hyperspherical front
  - hyperspherical GSF accelerate even more the rate of convergence (more natural in Ω_0)

- **TWO S-WAVE MODELS**

  1) **Analytical three-body Coulomb break-up model: HGSF gives excellent agreement.**
     USEFUL TOOL to test numerical methods (at any energy !)

  2) **Double ionization S-wave model for high electron impact energy :**
     should hopefully help in identifying what is going on in real (difficult) problem.
     Spherical and Hyperspherical Sturmian Functions: agree with each other
     → can other theories at least agree on this simplified problem?

This is a contribution towards elucidating …

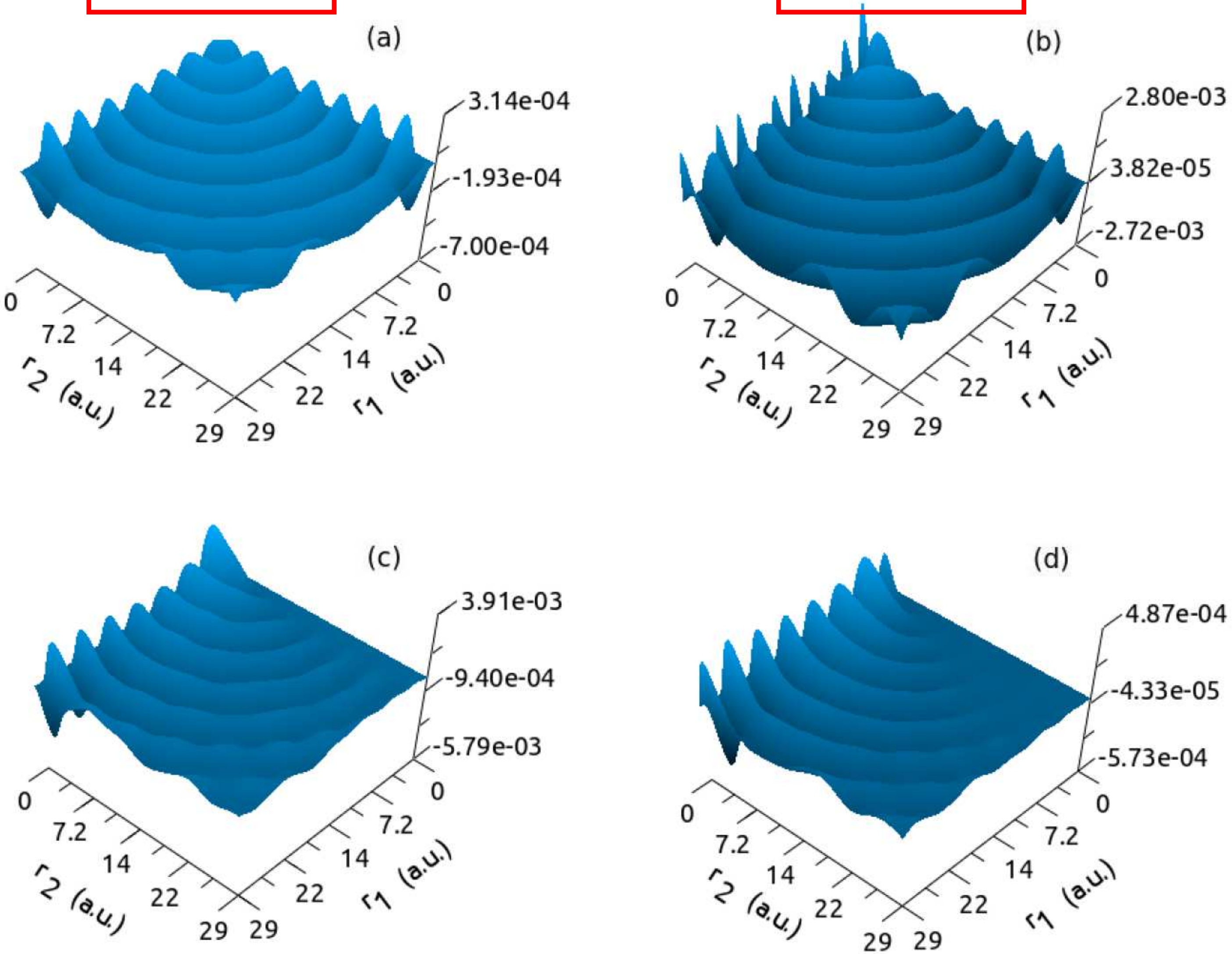
+ Preliminary results for the full (e,3e) process !

FULL
(e,3e)

All L

L=0
$(l_1,l_2)=(1,1)$

L=1
$(l_1,l_2)=(0,1)$



L=1
$(l_1,l_2)=(1,2)$

L=2
$(l_1,l_2)=(1,3)$

Thank you for your attention !