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# Perturbative and non-perturbative methods for electron-impact ionization

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*Los Alamos National Laboratory*

## Outline

- Background. Electron-impact ionization (EII); why do we care?
- What does perturbative and non-perturbative mean?
- Where does it matter?
- Examples of ionization of small targets – mostly atoms
- Ionization of larger, more complex targets.
- Plasma modeling – why we do all this!

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# Electron-impact ionization – what are we trying to do?

- We are trying to predict the cross sections for break-up of atoms and molecules by electron-impact, eg

$$e^- + He \rightarrow He^+ + e_1^- + e_2^-$$

  - The ionization probability is represented by a cross section – what can be measured
  - Cross sections can come in various forms:
    - Total cross section is just total probability for ionization
    - Differential cross section explores more of the details – cross section as a function of the angle and/or energy of the outgoing electrons

- We can solve directly the Schrodinger equation for such a system since all potentials are known – Coulomb terms in the Hamiltonian can be written down
  - Can't be solved analytically – "three-body problem"
  - Can be solved numerically – by making various approximations for the wavefunctions of the electrons and for the interaction terms in the Hamiltonian
  - Multi-electron targets require all electron-electron interactions to be considered

$$H_{\text{atom}} = -\frac{1}{2}\nabla_1^2 - \frac{1}{2}\nabla_2^2 - \frac{Z}{r_1} - \frac{Z}{r_2} + \frac{1}{|r_1 - r_2|}$$

# Theoretical approaches to EII: distorted-wave

- A distorted-wave (DW) approach treats the electron-electron interactions *perturbatively*
  - The incident, scattered, and ejected electronic wavefunctions are all 'distorted' by the potential of the target atom (including nuclear and electron potential terms)
  - One can choose the potential in which the wavefunctions are computed (eg 'post' or 'prior' forms of the interaction potential)
  - The interaction between the active electrons (incident and ejected) is often treated to **first-order** in perturbation theory
- Several sets of DW codes are available worldwide, such as LANL codes, FAC, etc.
- DW approaches are **much less computationally expensive** compared to close-coupling approaches
- They are accepted to be less accurate for neutral/near-neutral targets, but accuracy appears adequate for moderately and highly-ionized systems
- Fully relativistic DW versions are also available, as well as 'semi-relativistic' versions (often based on the Cowan code) in which one-electron terms are added into the non-relativistic Hamiltonian

# Theoretical approaches to EII: close-coupling

- Close-coupling approaches aim to treat the interaction between the outgoing (scattered & ejected) electrons in a non-perturbative manner in an effort to more accurately model the ionization process
  - **Non-perturbative** – the interaction between the active electrons is treated to all orders in perturbation theory.
- Several close-coupling approaches have been developed in recent decades:
  - Convergent close-coupling (CCC)
  - R-matrix (with pseudo states) (RMPS) and variants – notably B-spline R-matrix (BSR)
  - Time-dependent close-coupling (TDCC)
  - Exterior complex-scaling (ECS)
- **Each of these approaches has advantages/disadvantages**
  - **Most approaches are quite computationally expensive**
  - Fully relativistic versions of many of these approaches are also available
  - Accepted view is that one requires such close-coupled approaches for low-energy, neutral & near-neutral systems for which the electron-electron couplings are strong and cannot be treated perturbatively

# Theoretical approaches to EII: advantages/disadvantages

- **Perturbative/distorted-wave approaches**

- **Advantages**
  - Computationally fast
  - Can be straightforwardly combined with atomic structure codes to allow calculations for virtually any atom or ion
  - Convergence is straightforward to assess – usually through partial wave expansions
  - Can be easily extended to calculate ionization from inner shells, not just valence
  - Can be used to understand the physics of the scattering process
- **Disadvantages**
  - Not accurate for neutral targets and low incident energies
  - Are **especially** inaccurate when electron-electron interaction is dominant (eg when electrons are emitted with near-equal energies/angles
    - although various “fixes” such as Coulomb repulsion terms, such as Gamow factors, can be added

- **Nonperturbative approaches**

- **Advantages**
  - Should be **very** accurate – agreement with experiment for simple targets confirms this
  - Can produce all possible scattering processes in one calculation – ie all differential cross sections and all inelastic processes (in principle)
- **Disadvantages**
  - Computationally intensive
  - Numerical convergence can be sometimes challenging to confirm
  - Hard to implement for complex (open-shell) targets (eg Fe, W). Most methods restricted to (quasi) one- or two-electron targets, although R-matrix approach is an exception
  - Difficult to treat inner-shell ionization processes
  - “brute-force” calculation – does it allow for physics insight?

# Light atoms: total cross sections

- For H, He, & Li close agreement exists between theory & experiment
- Studies on Be also report good agreement between a variety of close-coupling methods
  - Recent study highlighted close agreement between RMPS, CCC, and TDCC methods
  - DW approaches ~ 20-30% higher for neutral atom but quickly approaches close-coupling results by two-times ionized
- Although studies of other light atoms (Z=1-10) are not comprehensive, where comparisons exist, agreement between different close-coupling approaches is generally satisfactory

Distorted-wave

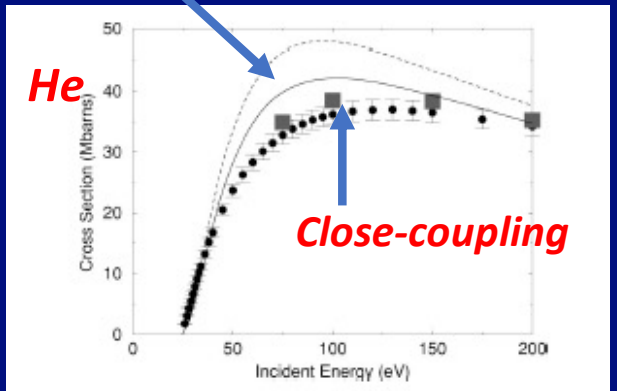


FIG. 1. Total electron-impact ionization cross section for helium. Solid squares: time-dependent close-coupling method, topped up at high angular momentum with distorted-wave results, solid curve: distorted-wave with exchange method, dashed curve: distorted-wave with no exchange method, solid circles: experimental measurements [18] (1.0 Mb = $1.0\times10^{-18}$ cm$^2$).

Distorted-wave

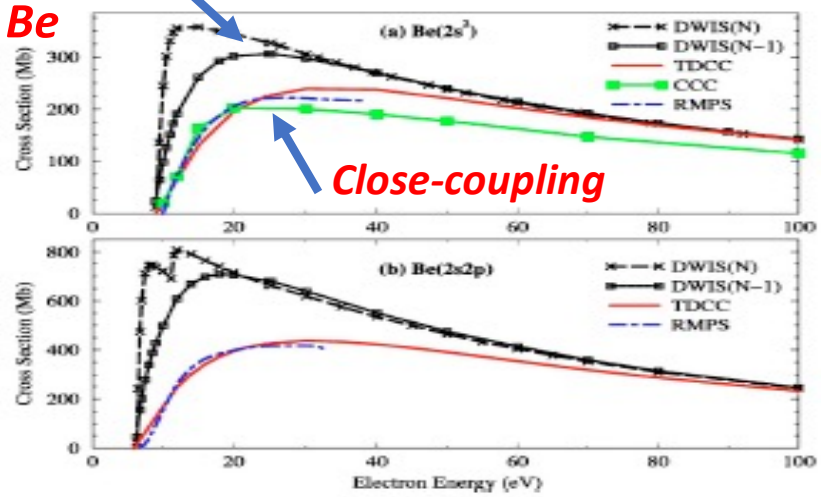


FIG. 1. Electron-impact ionization cross sections for neutral Be, from (a) the ground $1s^22s^2$ configuration and (b) the first excited $1s^22s2p$ configuration. The solid lines are the time-dependent close-coupling calculations. The dot-dashed lines are the RMPS calculations and the short-dashed (with crosses) and dotted lines (with squares) are the DWIS($N$) and DWIS($N-1$) calculations, respectively. The solid line with squares are CCC calculations from Ref. [7]. In (b) all calculations include ionization from both the $2s$ and $2p$ subshells. Also, the RMPS calculations are for ionization from the $1s^22s2p\ ^3P$ term only (1.0 Mb = $1.0\times10^{-18}$ cm$^2$).

*Pindzola & Robicheaux, PRA 61 052707 (2000)*
*Dipti et al, ADNDT 127, 1 (2019)*

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# Angular distributions: state-of-the-art comparison of He ionization

PHYSICAL REVIEW A **83**, 052711 (2011)

**Electron-impact ionization of helium: A comprehensive experiment benchmarks theory**

X. Ren,[1] I. Bray,[2] D.V. Fursa,[2] J. Colgan,[3] M.S. Pindzola,[4] T. Pflüger,[1] A. Senftleben,[1] S. Xu,[1] A. Dorn,[1,*] and J. Ullrich[1]
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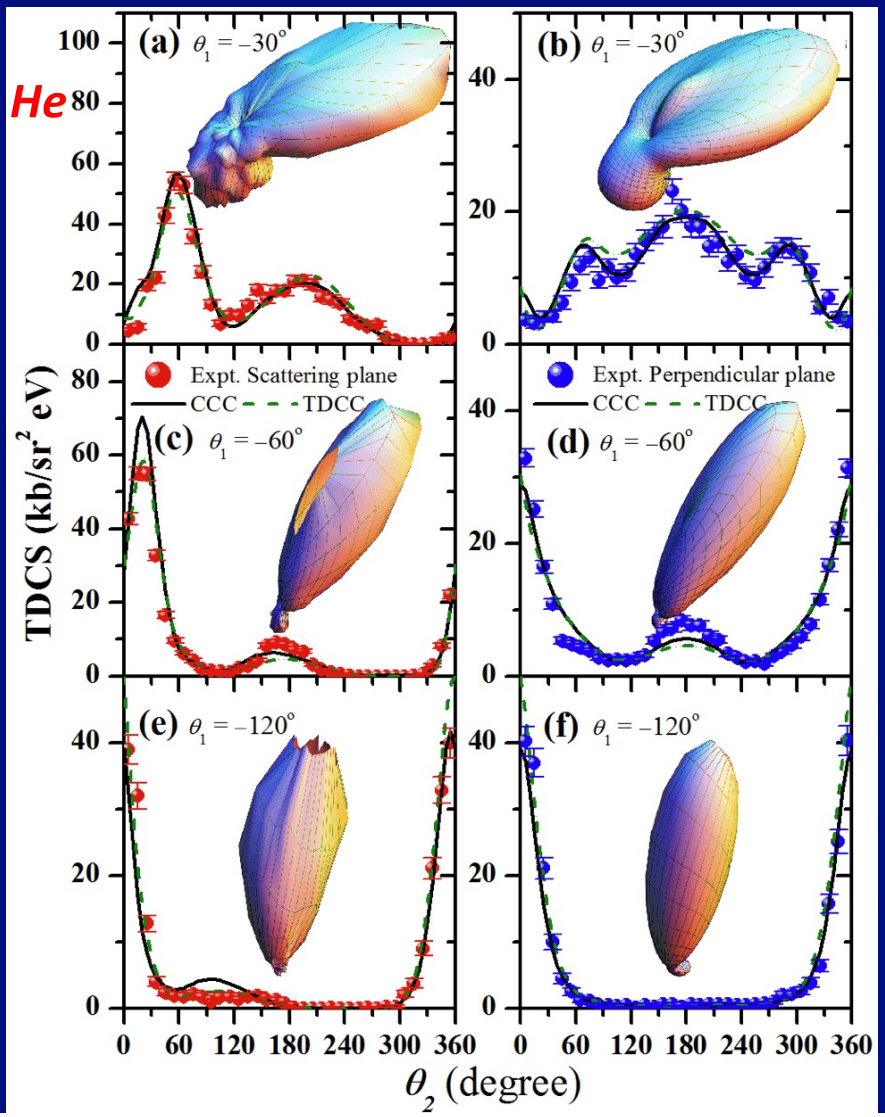
[2]*ARC Centre for Antimatter-Matter Studies, Curtin University, GPO Box U1987 Perth, Western Australia, Australia*
[3]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[4]*Department of Physics, Auburn University, Auburn, Alabama 36849, USA*
(Received 15 February 2011; published 24 May 2011)

- Measurements at MPI-K in Heidelberg have been able to measure the full angular distribution over most of the solid angle of ejection of the outgoing electron
- Agreement with non-perturbative methods (here, CCC and TDCC shown) is excellent
- The measurements and theory are ***absolute*** – close-coupling methods produce a fully consistent set of cross sections for all scattering processes in a given calculation.




***TDCS: triple differential cross section – ionization as a function of outgoing electron angle & energy***

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# Physics insight: Angular distributions from electron-impact ionization of He

PHYSICAL REVIEW A **92**, 052707 (2015)

**Propensity for distinguishing two free electrons with equal energies in electron-impact ionization of helium**

Xueguang Ren,[1,2,*] Arne Senftleben,[2,3] Thomas Pflüger,[2] Klaus Bartschat,[4] Oleg Zatsarinny,[4] Jamal Berakdar,[5] James Colgan,[6] Michael S. Pindzola,[7] Igor Bray,[8] Dmitry V. Fursa,[8] and Alexander Dorn[2]

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[5]*Institut für Physik, Martin-Luther Universität Halle-Wittenberg, D-06099 Halle/Saale, Germany*
[6]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
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(Received 27 August 2015; published 16 November 2015)

- Non perturbative calculations (RMPS as well as CCC and TDCC) were able to explain some features in the angular distributions and trace them to propensities for electron ejection

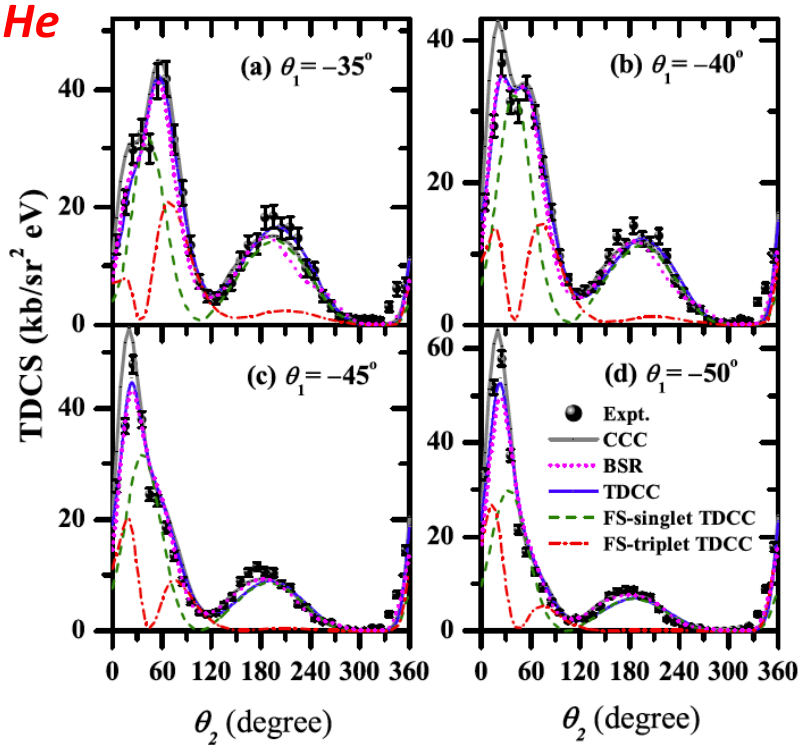


FIG. 3. (Color online) Experimental data compared with CCC, BSR, and TDCC predictions for the TDCS in the scattering plane ($E_1 = E_2 = 23$ eV) as a function of one-electron emission angle ($\theta_2$) with the other electron emission angle $\theta_1$ fixed to $-35^\circ$ (a), $-40^\circ$ (b), $-45^\circ$ (c), and $-50^\circ$ (d). Also shown are the TDCC results for the contributions from the final-state (FS) singlet and triplet spin channels constructed from coupling the spins of the two outgoing electrons. See text for details.

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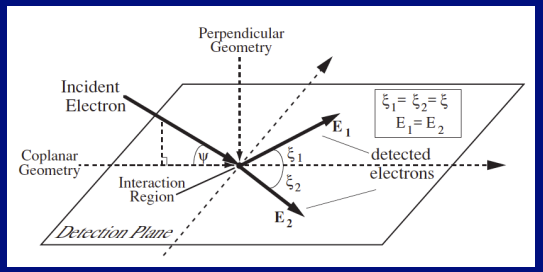
# Physics insights: Angular distributions and strong minima in TDCS of He

J. Phys. B: At. Mol. Opt. Phys. **42** (2009) 171001 (6pp) doi:10.1088/0953-4075/42/17/171001

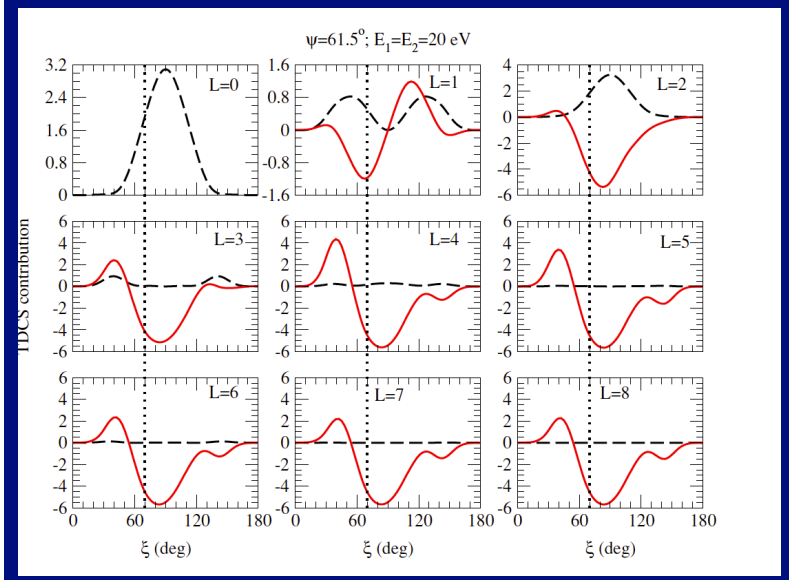
FAST TRACK COMMUNICATION

**Deep interference minima in non-coplanar triple differential cross sections for the electron-impact ionization of small atoms and molecules**

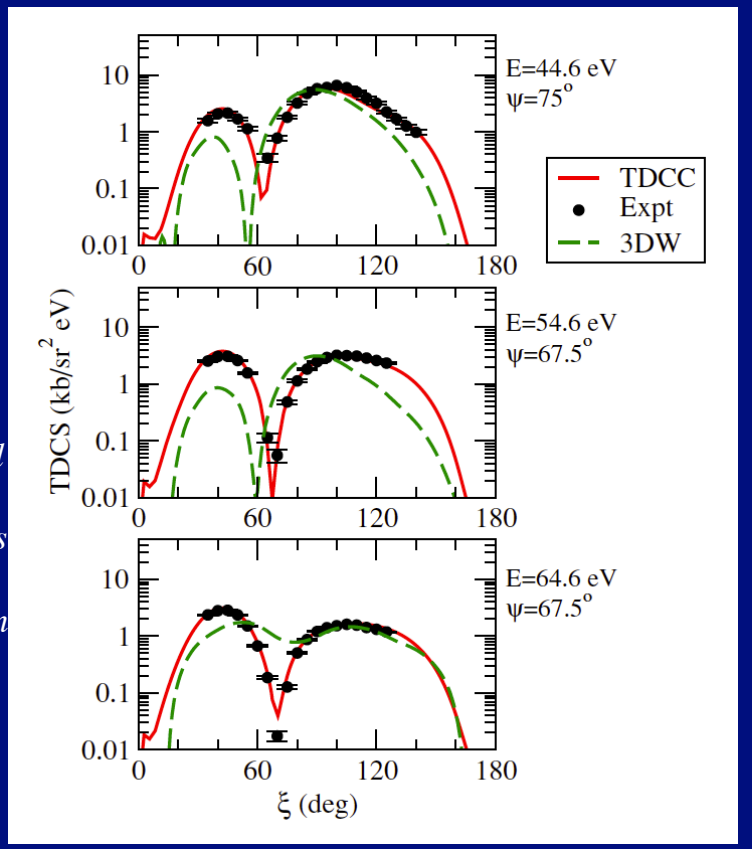
J Colgan[1], O Al-Hagan[2], D H Madison[2], A J Murray[3] and M S Pindzola[4]




- Measurements made in the 1990s showed an unexpected minimum in the angular distributions for electron ionization of helium
- Non perturbative & perturbative methods were used to study the physics of the scattering process
- A deep minimum was observed in both DW and TDCC calculations
- Interference between the partial waves in the quantum-mechanical scattering amplitude is related to the position of this minimum
- The minimum is also found for atomic H and molecular $H_2$!




*TDCC calculations showing the contributions from individual partial waves (dashed black lines) in each panel up to L = 8. The solid red lines signify the contribution from the interference (cross) terms inherent in the coherent sum in equation (1). For example, the red line in the upper right panel shows the contribution from the L = 0, 1, 2 cross terms. The vertical dotted lines indicate the position of the minimum in the TDCS.*

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# $H_2$ scattering – how do atoms and molecules differ?

nature physics

ARTICLES

PUBLISHED ONLINE: 16 NOVEMBER 2008 | DOI: 10.1038/NPHYS1135

## Atomic and molecular signatures for charged-particle ionization

Ola Al-Hagan[1], Christian Kaiser[2], Don Madison[1]* and Andrew James Murray[2]*

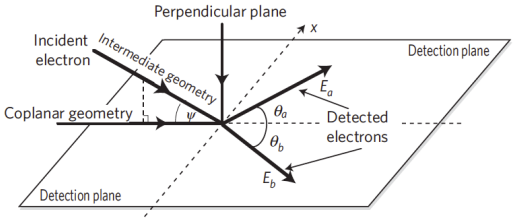
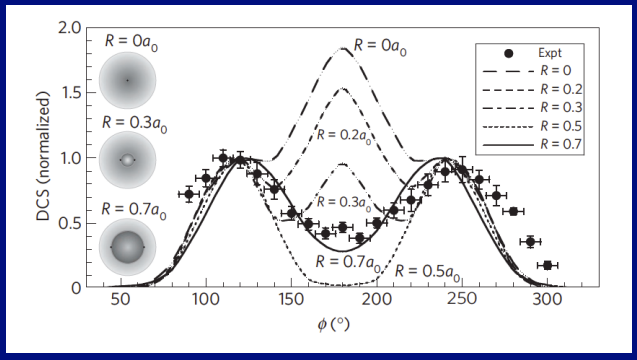
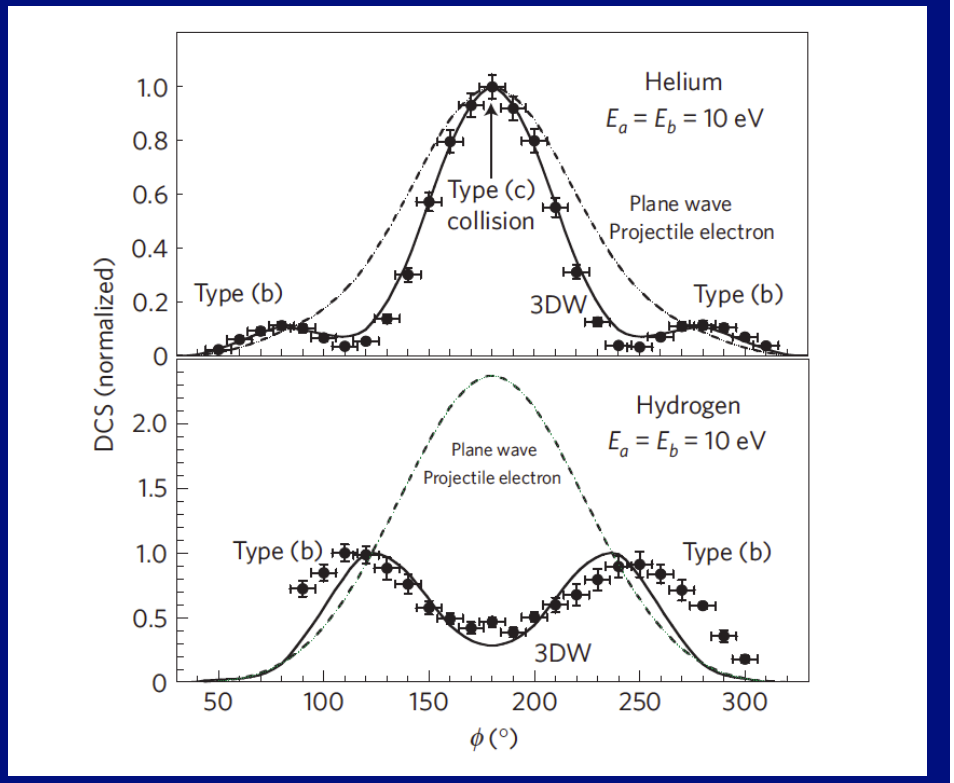



**Figure 1 | The experimental geometry.** A plane is defined by the detected electrons. The incident-electron gun can move from a coplanar geometry ($\psi = 0°$) to the perpendicular plane ($\psi = 90°$), where the angle $\varphi = \theta_a + \theta_b$ is defined. A common point between all planes occurs when $\theta_a = \theta_b = \pi/2$.

- Experimental studies of scattering from He and $H_2$ found significant differences for some geometries.
- In a special 'perpendicular' geometry, He angular distribution peaked at 180°, but $H_2$ had a dip there. ***Why***?
- Don Madison's **3DW** calculations (averaged over all molecular orientations), agreed with measurement, unlike plane-wave models. But what is the physics?

- The peak at 180° is caused by an electron scattering from the nucleus after a binary collision with an electron
- Scattering from a point charge (He) results in a different distribution than scattering from two centers ($H_2$), because the electron-nucleus scattering is stronger when a nucleus is present at the center-of-mass (as in He)
- So the 180° peak is stronger for atomic targets
- This implies scattering from tri-atomic molecules would produce a peak – and it does! $CO_2$ measurements have a peak at the center

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# $H_2$ scattering analysis – lots of physics to explore

PRL 101, 233201 (2008) PHYSICAL REVIEW LETTERS week ending 5 DECEMBER 2008

**Differential Cross Sections for the Ionization of Oriented $H_2$ Molecules by Electron Impact**

J. Colgan,[1] M. S. Pindzola,[2] F. Robicheaux,[2] C. Kaiser,[3] A. J. Murray,[3] and D. H. Madison[4]
[1]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[2]*Department of Physics, Auburn University, Auburn, Alabama 36849, USA*
[3]*School of Physics and Astronomy, University of Manchester, Manchester M13 9PL, United Kingdom*
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(Received 15 September 2008; published 2 December 2008)

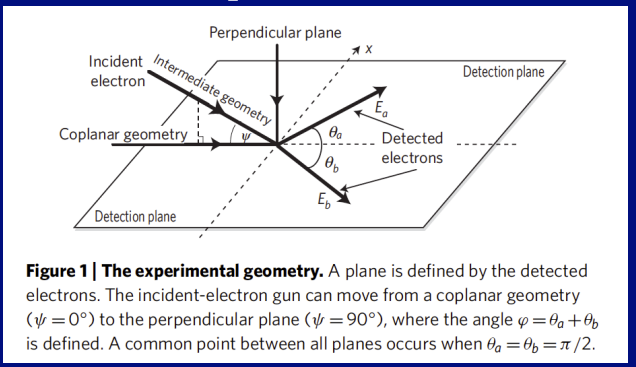
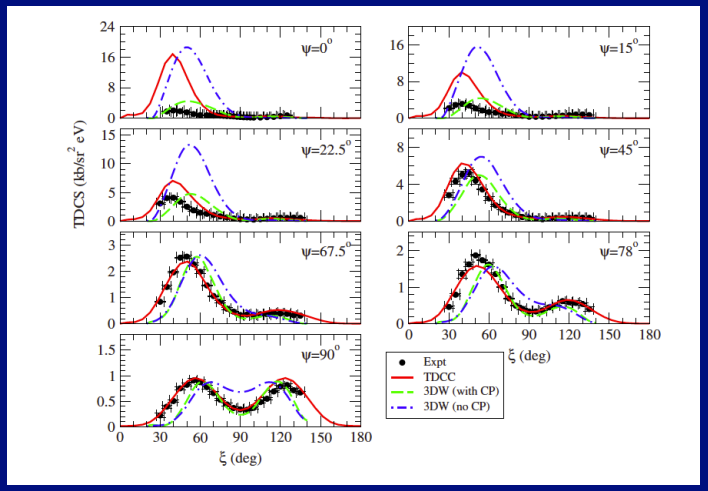
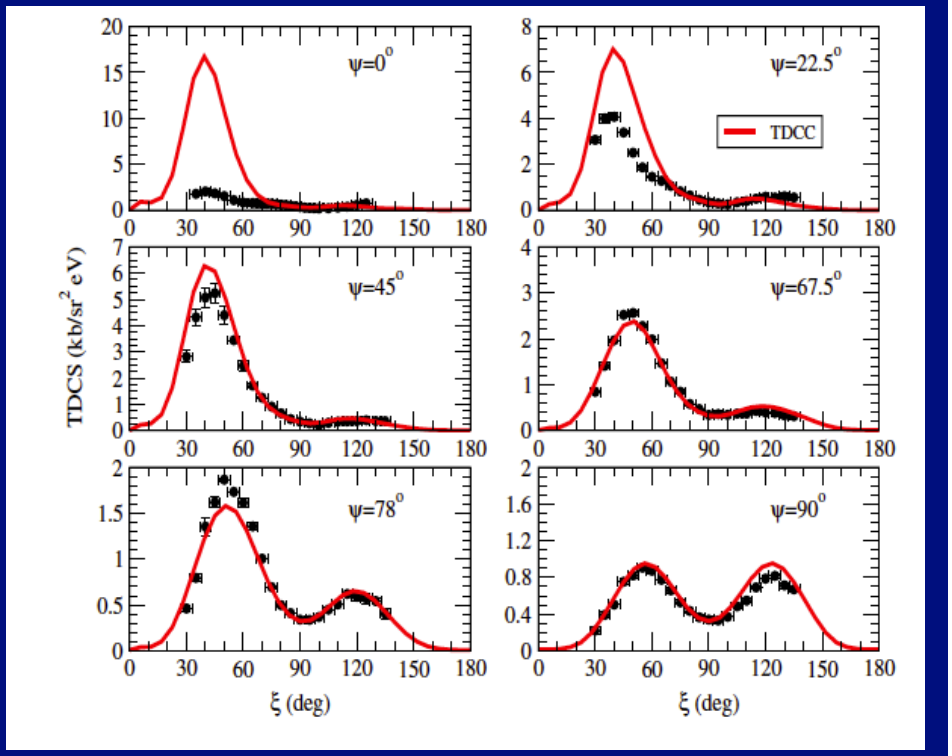



**Figure 1 | The experimental geometry.** A plane is defined by the detected electrons. The incident-electron gun can move from a coplanar geometry ($\psi = 0°$) to the perpendicular plane ($\psi = 90°$), where the angle $\varphi = \theta_a + \theta_b$ is defined. A common point between all planes occurs when $\theta_a = \theta_b = \pi/2$.

- A TDCC method for e-$H_2$ scattering was in development and I was able to compare with Andrew's measurements. This led to a fruitful collaboration with both Don Madison and Andrew Murray
- Agreement between TDCC and measurement was good for orientation-averaged cases.
- We were able to use TDCC (and 3DW, later) to examine the angular distributions from oriented $H_2$ molecules




***TDCC: time-dependent close-coupling method***

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# $H_2$ scattering analysis

PHYSICAL REVIEW A **81**, 030701(R) (2010)

**Electron-impact-ionization cross sections of $H_2$ for low outgoing electron energies from 1 to 10 eV**

Ola Al-Hagan,[1] A. J. Murray,[2] C. Kaiser,[2] J. Colgan,[3] and D. H. Madison[1]
[1]*Physics Department, Missouri University of Science and Technology, Rolla, Missouri 65409, USA*
[2]*School of Physics and Astronomy, Photon Science Institute, University of Manchester, Manchester M13 9PL, United Kingdom*
[3]*Theoretical Division, Los Alamos National Laboratory, New Mexico 87545, USA*
(Received 24 November 2009; published 29 March 2010)

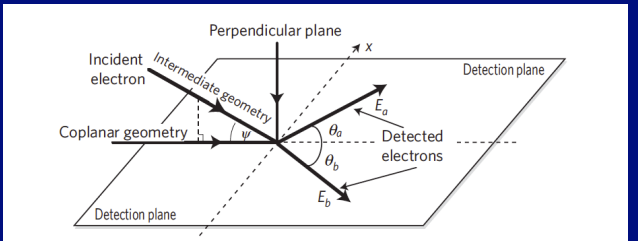
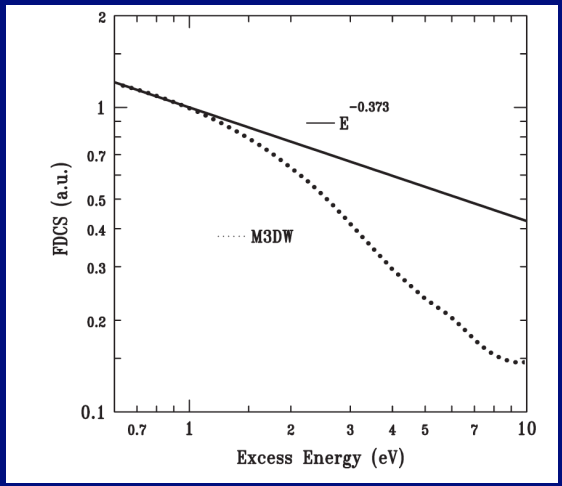
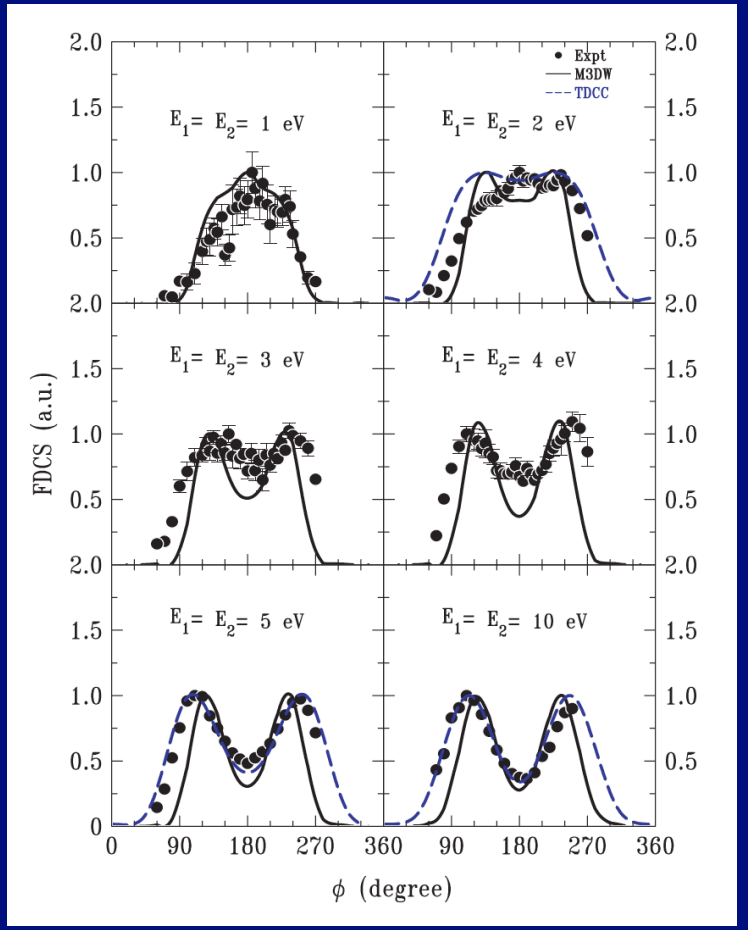



**Figure 1 | The experimental geometry.** A plane is defined by the detected electrons. The incident-electron gun can move from a coplanar geometry ($\psi = 0°$) to the perpendicular plane ($\psi = 90°$), where the angle $\varphi = \theta_a + \theta_b$ is defined. A common point between all planes occurs when $\theta_a = \theta_b = \pi/2$.

- Don's group worked on understanding the importance of PCI to these distributions. This is another example of Don leveraging the tools at his disposal to understand the scattering physics
- As the impact energy decreases (from 20 to 2 eV), the angular distributions change steadily in shape. Don showed that this was due to the increasing importance of PCI in the lower energy collisions. This is a *separate* effect from the electron-nucleus scattering found in the He and $H_2$ comparisons
- Don also related this to the threshold Wannier theory and was able to interpret his results to explore where this theory applies

***PCI: Post-collision interaction:*** *a measure of the correlation between the outgoing electrons. In the M3DW method, it is represented by a Ward-Macek Coulomb-distortion factor in the final-state wavefunction*

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# $H_2$ scattering analysis

PRL **109,** 123202 (2012) PHYSICAL REVIEW LETTERS week ending 21 SEPTEMBER 2012

**Strong Molecular Alignment Dependence of $H_2$ Electron Impact Ionization Dynamics**

X. Ren,[1] T. Pflüger,[2] S. Xu,[3] J. Colgan,[4] M. S. Pindzola,[5] A. Senftleben,[1] J. Ullrich,[1,2] and A. Dorn[1]

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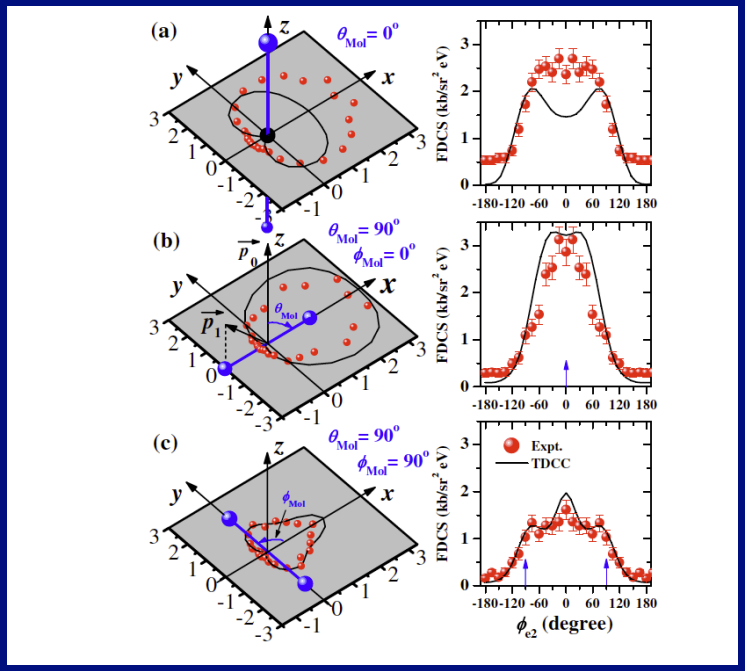
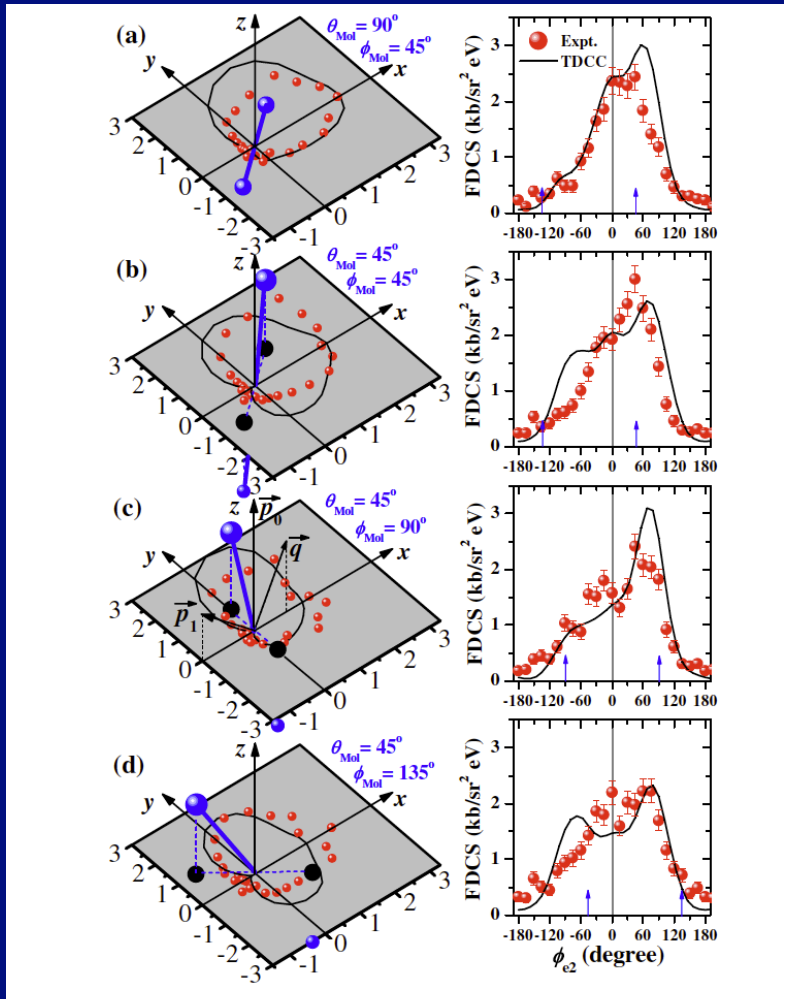
[2]*Physikalisch-Technische Bundesanstalt, Bundesallee 100, 38116 Braunschweig, Germany*
[3]*Institute of Modern Physics, Chinese Academy of Sciences, Lanzhou 730000, China*
[4]*Theoretical Division, Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[5]*Department of Physics, Auburn University, Auburn, Alabama 36849, USA*

(Received 2 August 2012; published 19 September 2012)

- There is lots of physics to explore in the physics of electron ionization of molecular $H_2$ – the molecular alignment at the time of ionization makes a difference to the resulting electron angular distribution patterns.
- Non perturbative calculations agree well (but not perfectly) with experiment

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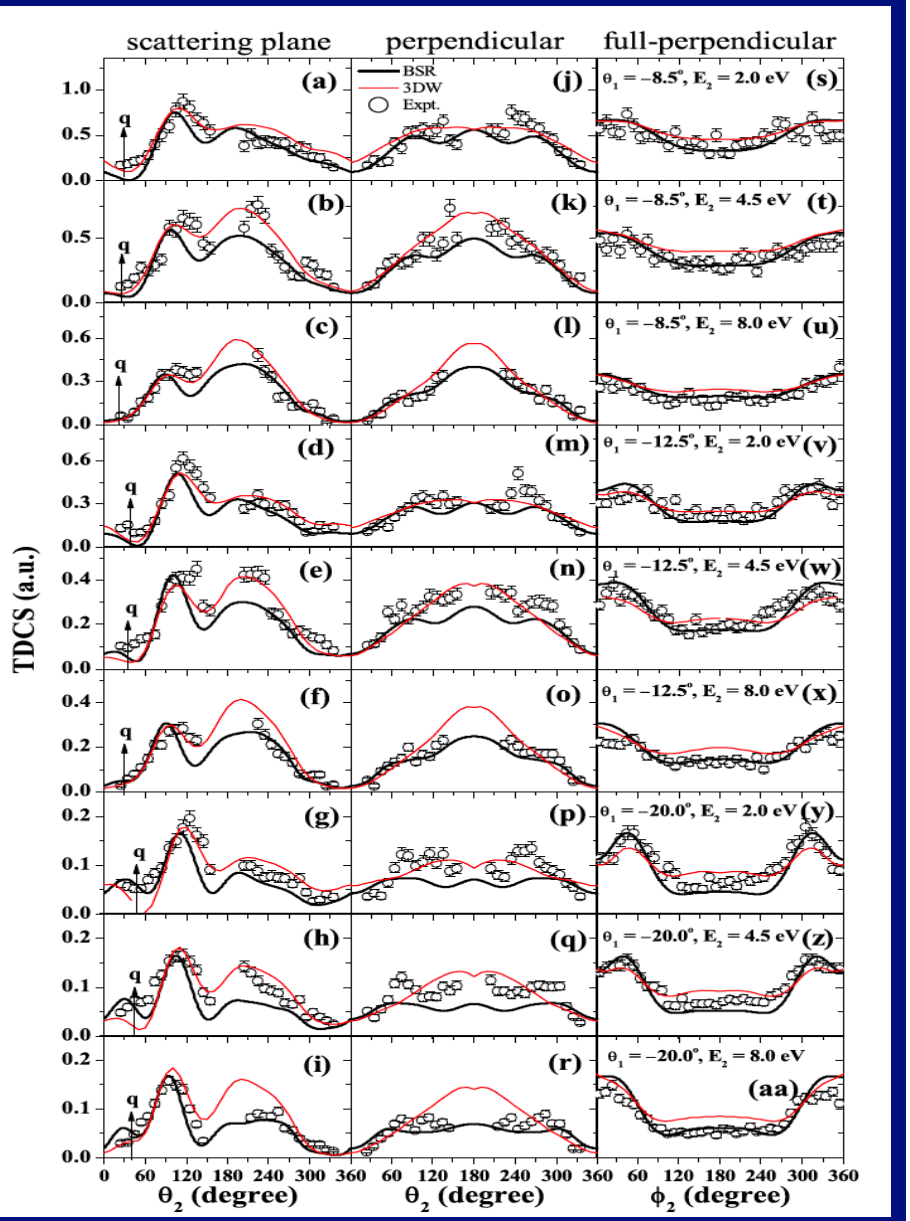
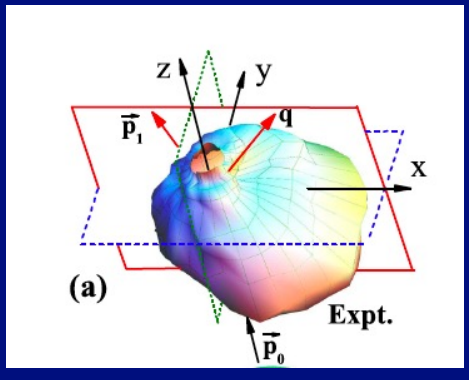
# Somewhat heavier atoms; neon

*Ne (Z=10)*

- TDCS measurements from neon were made by the Heidelberg group of Dorn et al
- They compared very well to two theoretical approaches: the **B-spline R-matrix** (BSR) approach of Bartschat and Zatsarinny, and a **distorted-wave (3DW)** approach of Don Madison and co-workers
- Comparisons were made for 3 slices through the 3-dimensional plane that the two outgoing electrons can occupy
- Various incident electron angles and energy sharings between the outgoing electrons were explored
- Both theories gave very good agreement with the data




*X. Ren et al, Phys. Rev. A 91, 032707 (2015).*

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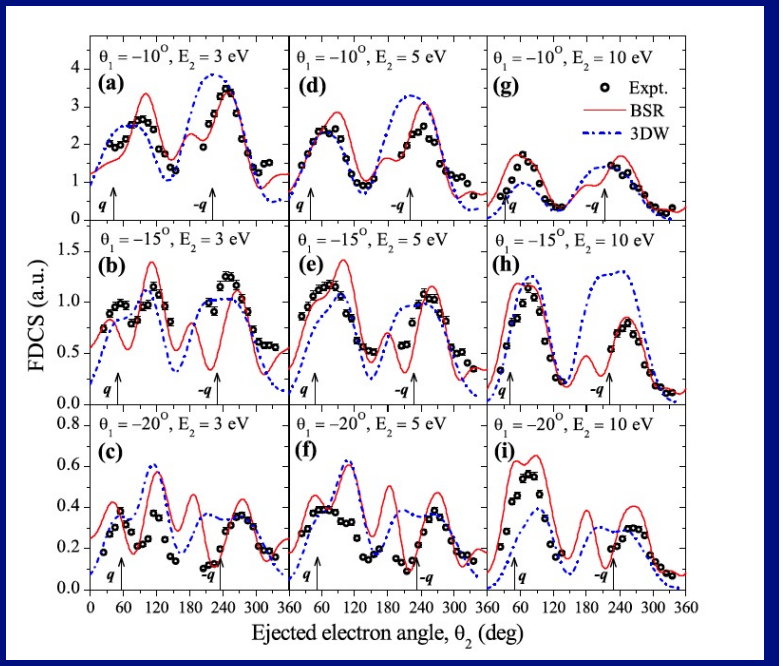
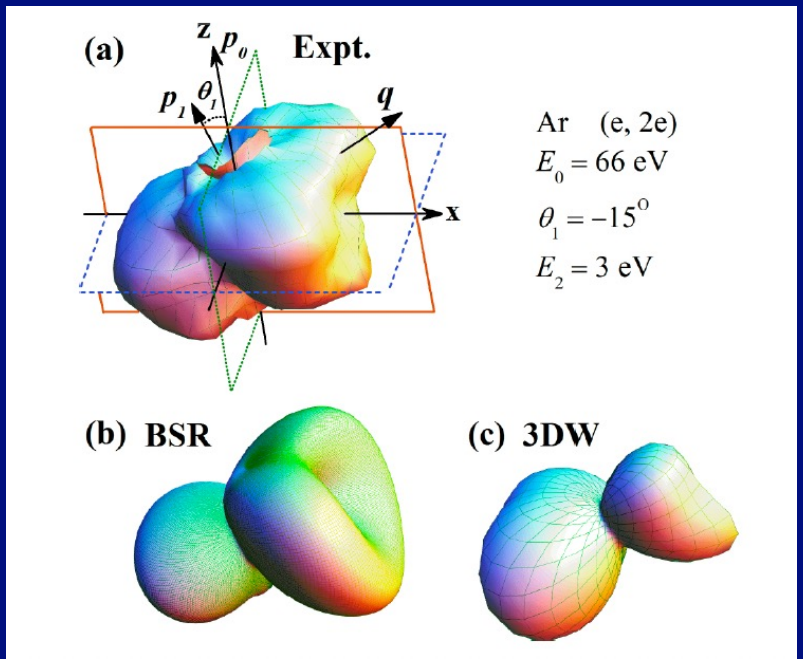
# Somewhat heavier atoms; argon

*Ar (Z=18)*

- Measurements were also then made on argon
- Agreement with the BSR approach was very good; the 3DW comparison was not quite as good
- Comparisons were again made for slices through the 3-dimensional plane that the two outgoing electrons can occupy
- As a heavier atom, it can be more difficult to describe the atomic structure correctly; also spin-orbit effects may be important

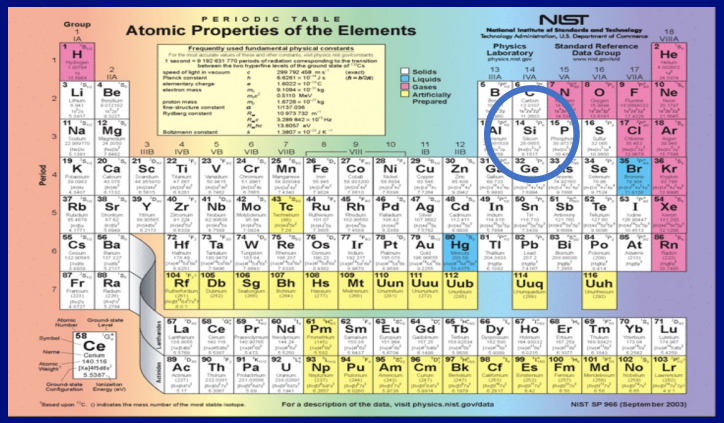



*X. Ren et al, Phys. Rev. A 93, 062704 (2016).*

# Moderately heavy atoms: Z~11-36

- For **total** cross sections, fewer close-coupling studies exist – some studies have been published for several noble gas atoms and quasi one-electron (Na) and two-electron (Mg) targets
- DW approach over-estimates the **absolute** cross section (compared to measurement) for neutral Si (not unexpected)
  - Although we note that at higher energies, the DW approach appears reasonable




*Si (Z=14)*

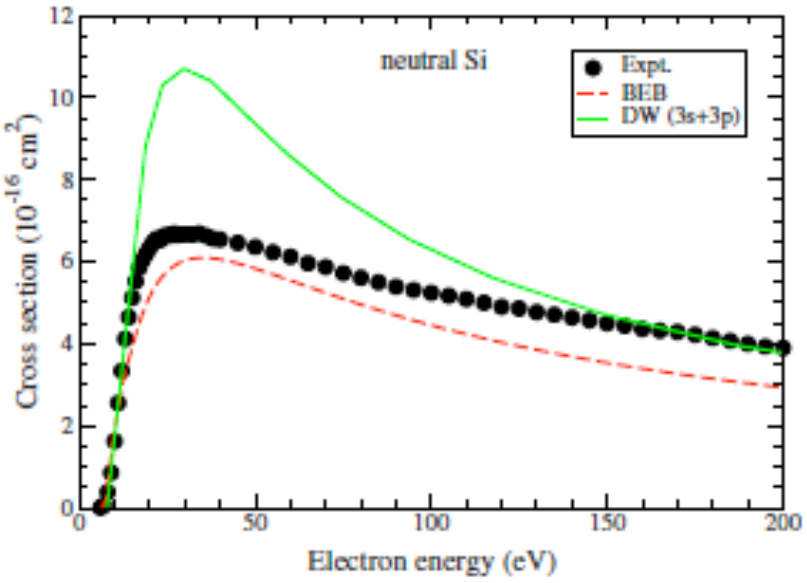
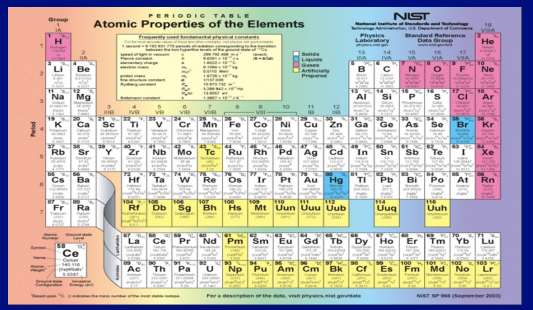



FIG. 2. (Color online) Electron-impact ionization cross sections for neutral Si. The partial cross sections from the $3s$ and $3p$ subshells of the ground $3s^2 3p^2$ configuration have been summed to compare with experiment. We compare our present distorted-wave calculations (DW) with the experimental measurements of Freund *et al.* [11] and binary-encounter-Bethe (BEB) calculations of Stone and Kim [32].

*Colgan et al, PRA 77, 062704 (2008)*

# Moderately heavy atoms: Z~11-36




- DW calculations for $Si^{2+}$ appear of acceptable accuracy
- And by $Si^{7+}$ DW appears very accurate

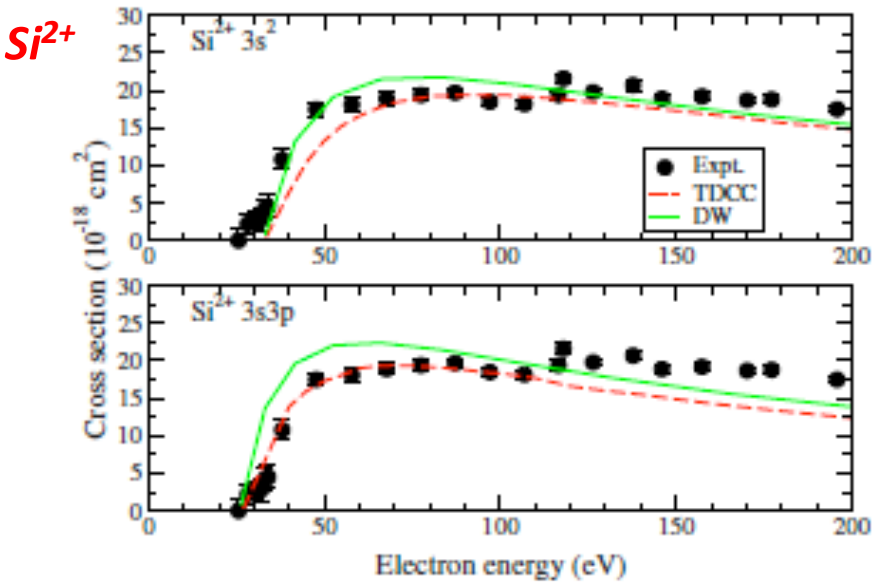



FIG. 3. (Color online) Electron-impact ionization cross sections for $Si^{2+}$ from the ground $3s^2$ and excited $3s3p$ configurations. In both cases, we compare our present distorted-wave calculations (DW) and time-dependent close-coupling (TDCC) calculations with the experimental measurements of Djurić *et al.* [12].

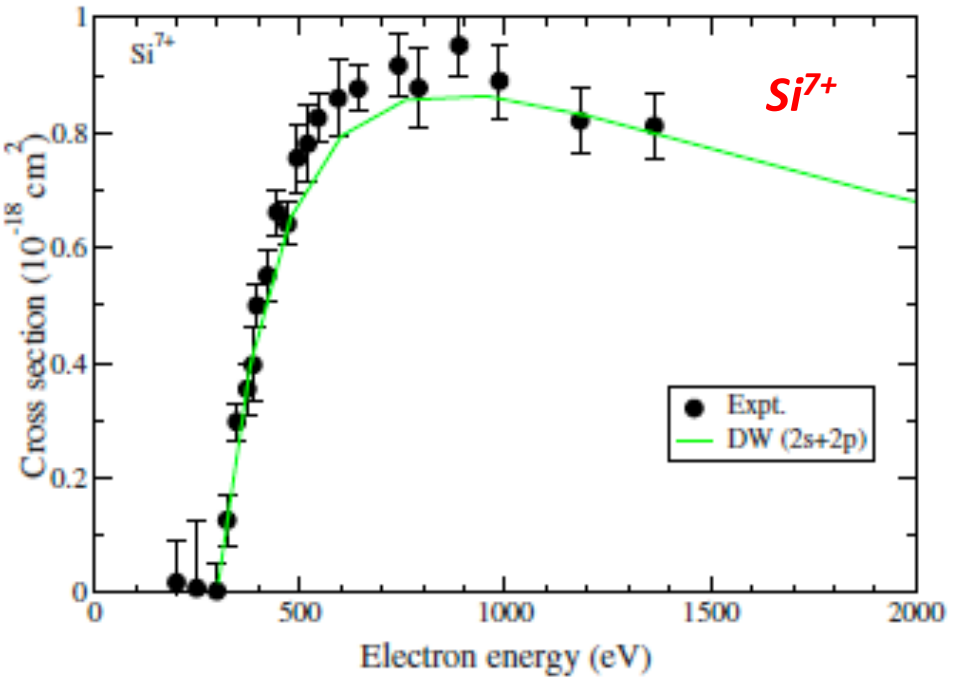
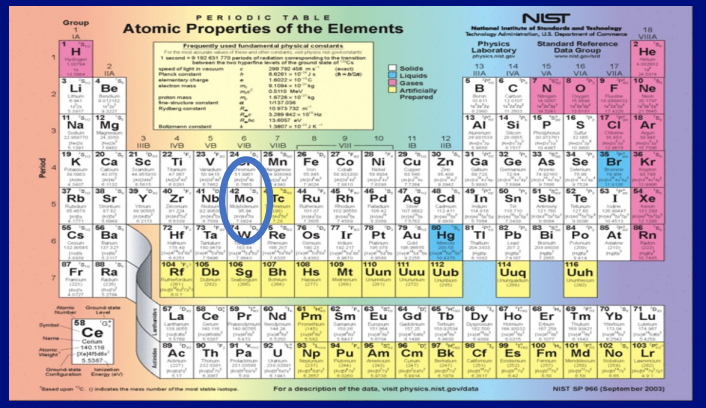



FIG. 5. (Color online) Electron-impact ionization cross sections for $Si^{7+}$. The partial cross sections from the $2s$ and $2p$ subshells of the ground $2s^22p^3$ configuration have been summed to compare with experiment. We compare our present distorted-wave calculations (DW) with the measurements of Zeijlmans van Emmichoven *et al.* [15].

# Moderately heavy atoms: Z~11-54




- Transition metals?
  - Not aware of (m)any close-coupling calculations
- Close-coupling approaches generally have difficulty with targets in which the atomic structure is complex
  - TDCC, ECS, and CCC are restricted to (at best) quasi one-electron and two-electron targets
    - And even these calculations may involve approximations with respect to structure
  - R-matrix approach **can** be applied to complex targets, but convergence can be difficult
  - Relatively few ionization calculations have been performed with R-matrix calculations for heavy atoms – excitation is in some sense easier
  - Preliminary calculations were performed on $Mo^+$ in 2005
    - Correlation and term-dependence in the initial state was explored

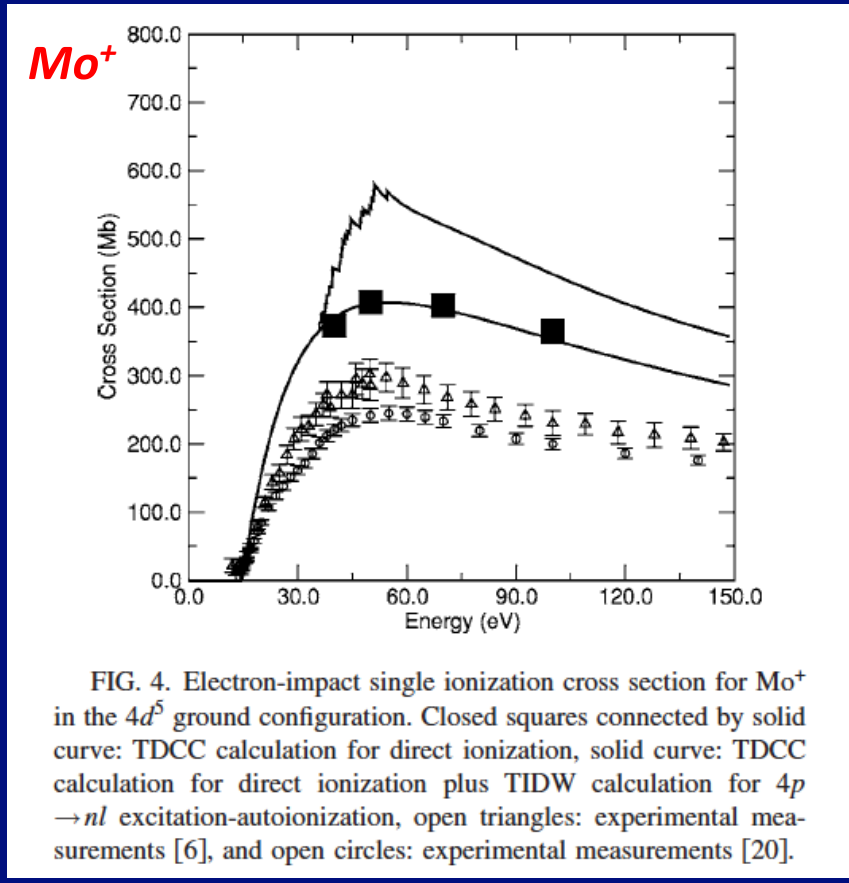



FIG. 4. Electron-impact single ionization cross section for $Mo^+$ in the $4d^5$ ground configuration. Closed squares connected by solid curve: TDCC calculation for direct ionization, solid curve: TDCC calculation for direct ionization plus TIDW calculation for $4p \rightarrow nl$ excitation-autoionization, open triangles: experimental measurements [6], and open circles: experimental measurements [20].

*Ludlow et al, PRA 72 032729 (2005)*

# Electron-impact ionization of W: comparison with measurement

- FAC code also used to investigate single ionization of $W^+$ ions
  - Sensitivity noted of the cross section to the choice of local central potential within the FAC calculation
  - Authors used potentials arising from either
    - $W^+$ [$5d^46s + 5d^5 + 5d^36s^2$ ] or
    - $W^{2+}$ [$5d^36s + 5d^4 + 5d^26s^2$ ]
  - Different cross sections obtained depending on this choice
  - Underlines the difficulty even of the structure calculations that are required for such complex ions
  - Excitation-autoionization is also expected to contribute to the cross section in this case
  - A further complication is that the distorted-wave approximation may also be inaccurate for such low-charged ions

$W^+$

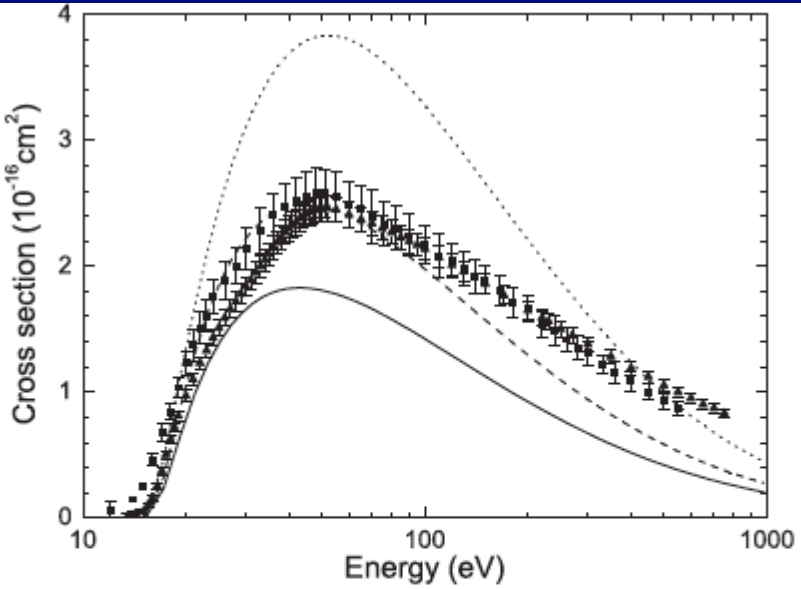



**Fig. 1.** DI cross section of a 5*d* and 6*s* electron for the ground level $5d^46s^6D_{1/2}$ of $W^+$ forming $W^{2+}$. Different curves represent various configurations for the optimization of the single local central potential: dotted curve, the configurations $5d^46s+5d^5+5d^36s^2$ of $W^+$; dashed curve, the configurations $5d^4+5d^36s+5d^26s^2$ of $W^{2+}$; solid curve, the configurations $5d^4+5d^36s+5d^26s^2+5d^25f^2+5d5f^26s+5f^26s^2+5d^25g^2+5d5g^26s+5g^26s^2$ of $W^{2+}$. The filled rectangles [5] and triangles [10] are the experimental results.

*There has been some Dirac R-matrix calculations of electron-impact excitation in recent years by the Belfast/Auburn groups.*

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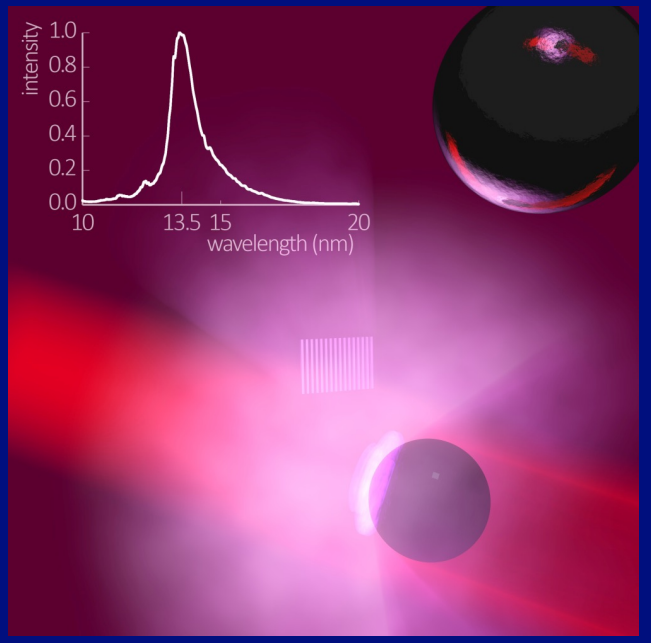
*Zhang & Kwon, Int. J. Mass. Spect. 356, 7 (2013)*

# *Putting this all together – ionization cross sections are needed in plasma modeling*

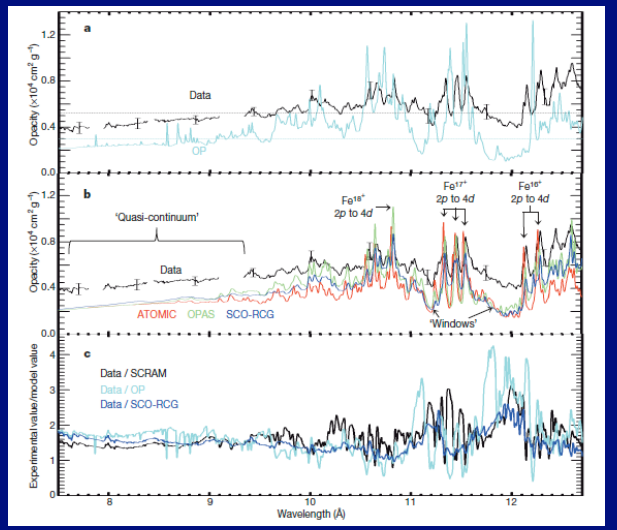
- **Total** ionization cross sections are needed to perform collisional-radiative modeling of partially-ionized plasmas
  - Other inelastic cross sections also required – excitation and inverse processes
  - One requires cross sections from all states of all ions in the plasma – data management can be an issue
  - Examples include atmospheric plasma modeling, and Sn laser-produced plasmas used in lithography

- In radiation-dominated plasmas, **photo-processes** are also needed (photoionization and photoexcitation)
  - A notable example is in the modeling of our Sun, where the photoionization cross sections have recently been questioned
  - In this case (mid-ionized Fe), the perturbative and non-perturbative cross sections are in good agreement

**Non-LTE** modeling of Sn plasmas is a key component in understanding the plasmas in EUV lithography




**LTE** modeling of Fe plasmas is key to some of the puzzles remaining in solar modeling

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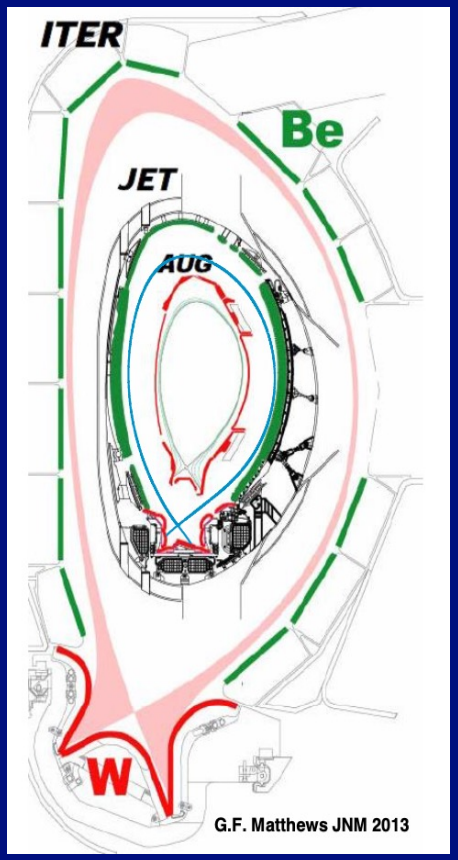
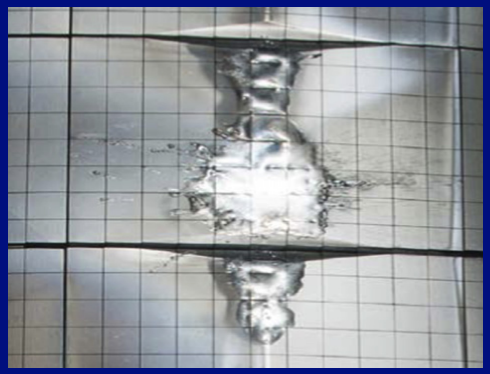
# *Angular* distributions of ionized electrons are also important for modeling magnetically confined plasmas

- **Disruption** in a tokamak plasma:
  - One of the largest uncertainties of the tokamak confinement concept.
  - Disruption event caused by plasma instabilities, etc.
  - Quenches plasma from 5-10 keV to 1-10 eV over a ms. No fusion.
  - Leads to a chain of events that critically damages vessel.
  - We need to safely be able to “turn off” the plasma to mitigate damage.
  - A way to turn “off” the plasma is to inject a mid/high Z element that will act as a coolant, since the higher Z elements radiate power much more efficiently than low Z species.
  - However, these species can & will ionize in the plasma, and the resulting electrons can “runaway” – get accelerated by the plasma fields to extremely high energies. To mitigate this, we need to know where the electrons move and with how much energy. **Atomic physics can help**!



***Our goal is to provide accurate angular distributions from a variety of relevant species to inform plasma kinetic simulations***

# *Macroscopic codes often neglect microscopic physics*

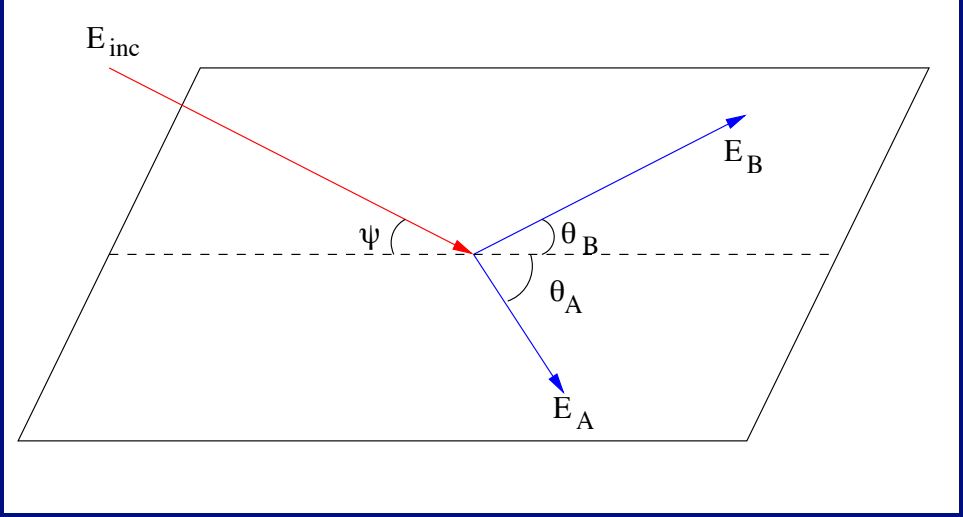
**Atomic Description → Bulk and Transport Properties → Macroscopic Behavior**

Often, Monte Carlo codes don't include much atomic physics – in particular, they assume that ionized electrons are ejected isotropically or in the forward direction.

– Is this approximation valid? If not, how much difference would these angular distributions make to the conclusions of the Monte Carlo simulations?

– Hence the need for accurate angular distributions.

$$e^-(\varepsilon_{\text{inc}}) + Z^{q+}(l) \longrightarrow e^-(\varepsilon_s, \Omega_s) + e^-(\varepsilon_{\text{ej}}, \Omega_{\text{ej}}) + Z^{(q+1)+}(u)$$



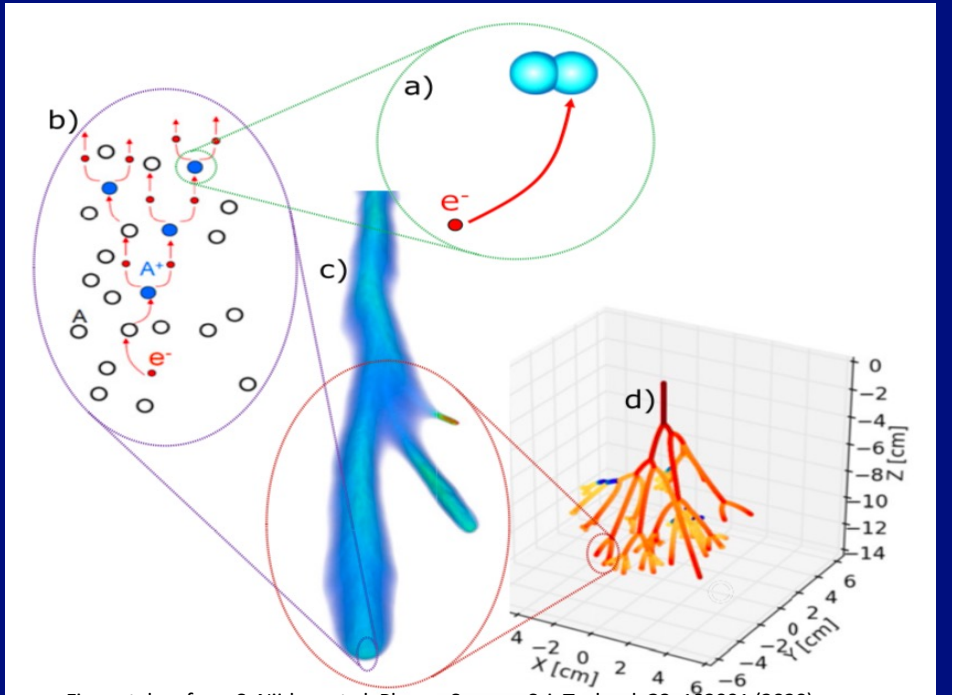




Figure taken from S. Nijdam et al. Plasma Sources Sci. Technol. **29**, 103001 (2020)

***Our goal is to provide accurate angular distributions from a variety of relevant species to inform plasma kinetic simulations***

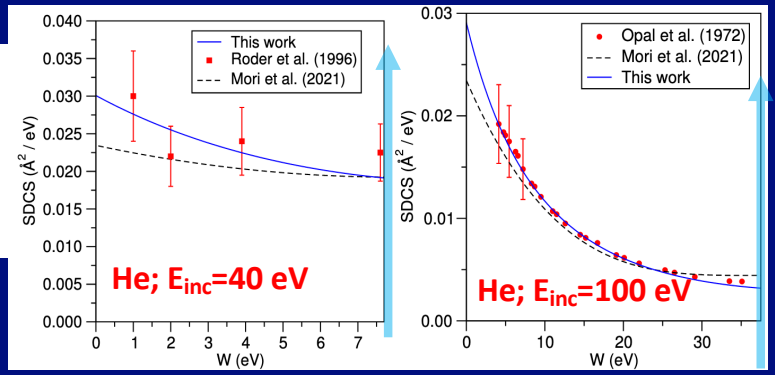
# Electron-impact ionization collision model for Monte Carlo kinetic simulations

*For more details see Ryan Park's talk on Tuesday: GT2.00005*

$$e^-(\varepsilon_{inc}) + Z^{q+}(l) \longrightarrow e^-(\varepsilon_s, \Omega_s) + e^-(\varepsilon_{ej}, \Omega_{ej}) + Z^{(q+1)+}(u)$$

## Electron energy-sharing:

40- and 100-eV electron-He single differential cross sections. Figure taken from P. Garkoti et al. Atoms **10**, 60 (2022)




**Collision models in MC simulations generally assume:**
**Equal-energy sharing ($E_s$=$E_{ej}$)**

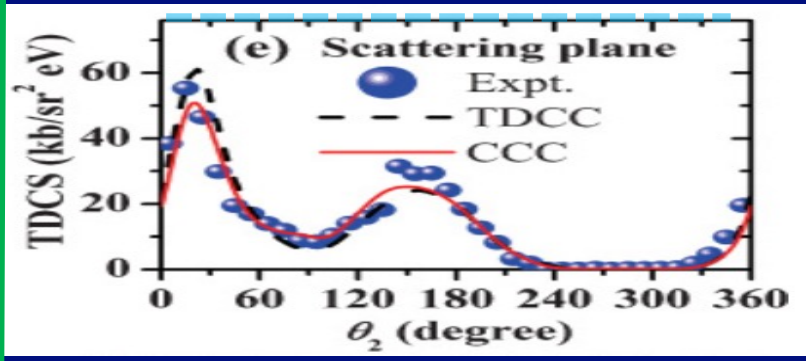
$$\frac{d\sigma(\varepsilon_{inc})}{d\varepsilon_{ej}} = \sigma(\varepsilon_{inc})\Sigma(\varepsilon_{inc}, \varepsilon_{ej}) \qquad \int_0^{(\varepsilon_{inc}-\varepsilon_{ion})/2} \Sigma(\varepsilon_{inc}, \varepsilon_{ej})d\varepsilon_{ej} = 1$$

$$R(\varepsilon_{inc}, \varepsilon_{ej}) = \int_0^{\varepsilon_{ej}} \Sigma(\varepsilon_{inc}, \varepsilon'_{ej})d\varepsilon_{ej'}$$

$$\varepsilon_{ej}(R, \varepsilon)$$

$$\frac{d\sigma(\varepsilon_{inc})}{d\varepsilon_{ej}d\Omega_s d\Omega_{ej}} = \frac{d\sigma(\varepsilon_{inc})}{d\varepsilon_{ej}} I(\varepsilon_s, \Omega_s) I(\varepsilon_{ej}, \Omega_{ej})$$

## Angular distribution of electrons:




40.6-eV electron-He triple differential cross section. Figure taken from X. Ren Phys. Rev. A **82**, 032712 (2010).

$E_1$ = 4 eV,
$\theta_1$ = -70 deg.
$E_2$ = 20 eV

**Collision models in MC simulations generally assume:**
**Isotropic scattering, I = 1 / 4$\pi$**

$$\int I(\varepsilon, \Omega)d\Omega = 2\pi \int_0^{\pi} d\theta \sin(\theta) I(\varepsilon, \theta) = 1$$

$$R = 2\pi \int_0^{\theta_s} I(\varepsilon_s, \theta'_s)d\theta'_s$$

$$\cos(\theta_s) = \mathcal{F}(\varepsilon_s, R, \ldots)$$

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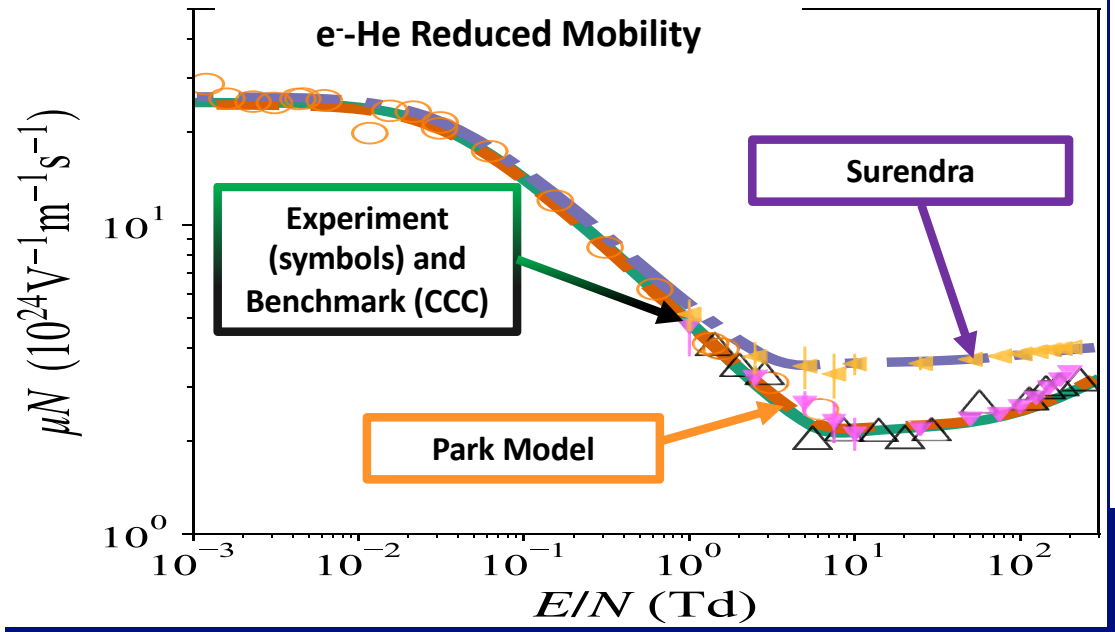
# We have developed a new anisotropic angular distribution functions

$$I(\varepsilon,\Omega)=\frac{1}{4\pi}\frac{\varepsilon}{\left[1+\varepsilon\sin^2(\theta/2)\right]\ln(1+\varepsilon)}$$

Park et al. Plasma Sources Sci. Technol. **31**, 065013 (2022)

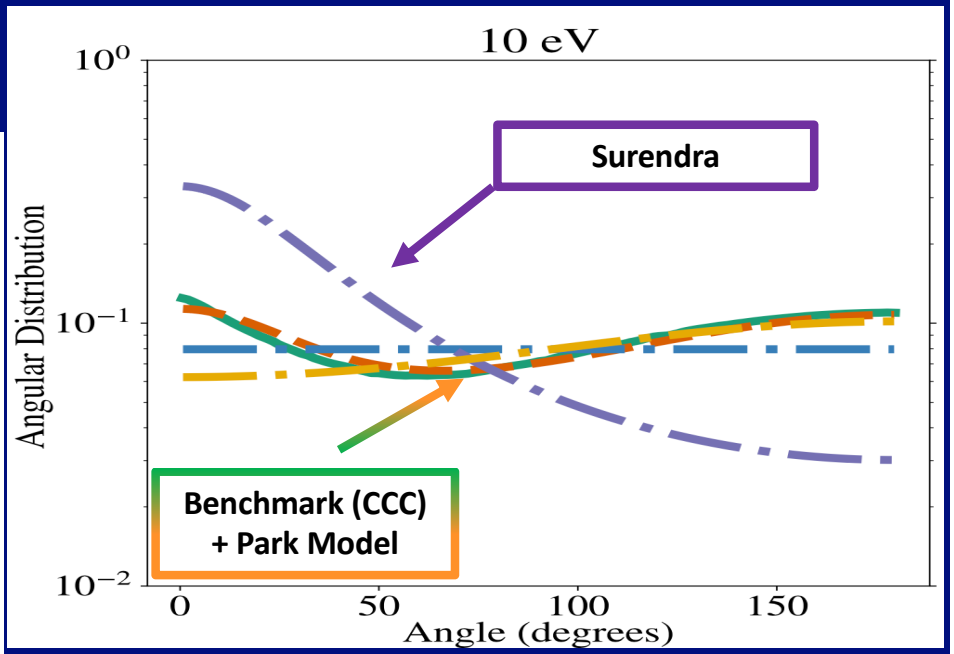
$$I(\varepsilon,\Omega)=C(\varepsilon)\frac{\eta_F(\varepsilon)(\eta_F(\varepsilon)+1)}{\pi\left[2\eta_F(\varepsilon)+1-\cos(\theta)\right]^2}+\left[1-C(\varepsilon)\right]\frac{\eta_B(\varepsilon)(\eta_B(\varepsilon)+1)}{\pi\left[2\eta_B(\varepsilon)+1+\cos(\theta)\right]^2}$$

***For more details see Ryan Park's talk on Tuesday: GT2.00005***




0-D kinetic simulations of the swarm transport parameters for He. The Park et al. Plasma Sources Sci. Technol. **31**, 065013 (2022) model simulations are in excellent agreement with experiment without the need to adjust the data.

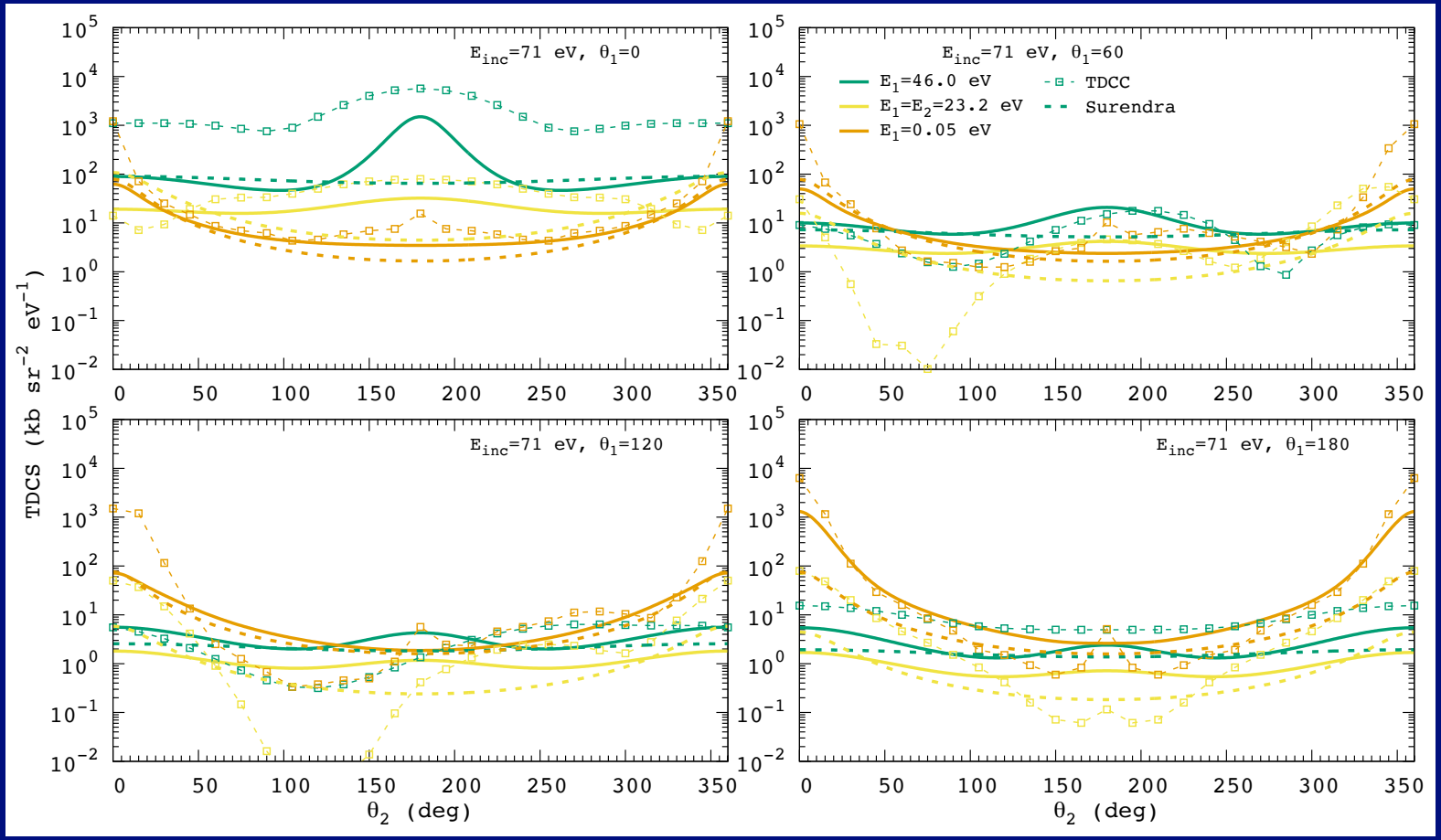
**Elastic: e⁻ + He**




Electron elastic scattering from He. Analytic anisotropic scattering models of Park and Surendra are compared with benchmark.

# 71 eV electron-He triple-differential cross sections: New preliminary model

***How do our calculations compare to what is used currently in PIC codes?***




**Squares: TDCC**
**Dotted-lines: Surendra model:**
**Solid lines: *new* model:**

$$I(\varepsilon,\Omega)=\frac{1}{4\pi}\frac{\varepsilon}{\left[1+\varepsilon\sin^2(\theta/2)\right]\ln(1+\varepsilon)}$$

$$I(\varepsilon,\Omega)=C(\varepsilon)\frac{\eta_F(\varepsilon)(\eta_F(\varepsilon)+1)}{\pi\left[2\eta_F(\varepsilon)+1-\cos(\theta)\right]^2}+\left[1-C(\varepsilon)\right]\frac{\eta_B(\varepsilon)(\eta_B(\varepsilon)+1)}{\pi\left[2\eta_B(\varepsilon)+1+\cos(\theta)\right]^2}$$

# Conclusions: A suite of methods are available in atomic physics to compute collisional cross sections

**Perturbative** (eg distorted-wave) methods have many advantages; they are

- Accurate for high electron energies
- Accurate for mid and highly ionized ions
- Computationally relatively fast to compute
- It is fairly straightforward to compute ionization from inner-shell states

But..

- They are **not accurate** for low energies and neutral/near-neutral atoms (or molecules)
- This implies DW methods are most suitable for “hot” plasmas where most ions are moderately or highly ionized

**Non-perturbative methods** are useful for neutral/near-neutral systems

- They are **very** accurate when converged because they contain most of the key physics describing the interactions of the incoming and ionized electron.
- However, they have some drawbacks:
  - They are computationally intensive
  - They can be  difficult to implement for open-shell systems – R-matrix methods have the best hope
  - Most useful for “low-temperature” plasma modeling

Which method you want to use will depend on what application you care about – ie, what plasma conditions are relevant!

In all cases, atomic & molecular collisional cross sections remains a key quantity in modeling a wide variety of industrial and astrophysical plasmas

# Perturbative and non-perturbative methods for electron-impact ionization

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LAUR-23-31245

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**Missouri University of S&T: Don Madison**

**MPI Heidelberg – Alexander Dorn**

**Drake University – Klaus Bartschat**