Current Research and Future Plans

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PRL, Ahmedbad

At IISER Pune, Jan 2014
Broad Areas

Dissociative Ionisation

Photoionisation of CS$_2$

Summary

What Next?

Plans for IISER-Pune
Kinematically complete dissociative ionisation

- Major part: high energy electron-impact on molecules
- Technique: ion-momentum spectrometry/coincidence momentum imaging
- Successes: identification of bent states of CO$_2^+$ and determining the lifetimes of metastable states
- Similar experiments using photons (Indus-1) and fast high charge ions (IUAC)
- Inverse kinematics is probably the only way to access the dynamics of transient molecular ions in conjunction with the computation of PE surfaces

Production of clusters (single/mixed species) with a goal to study heterophase photochemical reactions

- cluster production by sputtering and laser ablation, mass spectrometric detection
- only minor success overall

State selective dissociative ionization

- electron–ion multi-fold differential cross-sections
  [correlated momentum maps]
- under photo-absorption (as well as ion impact)
Instruments built over the last few years

- Ion momentum spectrometers with coincidence momentum imaging
- Time-of-flight mass spectrometer
- Ion sputtering cluster source, laser ablation cluster source, supersonic nozzle beam cluster source
- Cylindrical Retarding Potential analyser (space instrumentation)
- A combined Cylindrical Mirror Analyser for electrons and Ion Momentum Spectrometer
Dissociative Ionisation

Complete Kinematics

\[ \text{B} + \text{A} \rightarrow (\text{A} + \text{B})^{2+} + \text{B}^+ + \text{A}^+ \]

▶ No exact solutions to the multi-electron Hamiltonian; only approximate calculation of the transition matrix

▶ Measurement of DCS enhances our understanding of the dynamics

▶ Fragment kinematics leads to properties of the transient state

▶ Experimental Challenge: detect all fragments and determine the correlated kinematics of all fragments – ions, atoms and electrons

▶ Additional consideration: the transient state is not unique

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Dissociative Ionisation
Selectivity in excitation

Dissociation patterns expected to depend on which molecular state is accessed by the excitation. Can we control or select the excited state that participates in the DI process? Such selectivity will bring us closer to measuring the fully differential cross-section to enable a direct comparison with theory. Selectivity can be brought in by obtaining correlated ion momentum distributions in conjunction with ejected electron energies.
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Dissociative Ionisation

\[ AB \rightarrow AB^{n+} + 2e^- \rightarrow \text{fragments} + 2e^-, \ n \geq 2 \]
Dissociative Ionisation Strategy

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- Crossed photon and molecular beams, single collision conditions
- Ion momentum spectrometer augmented with the CMA
- Electrons of specific energy, selected by the CMA, start the ion TOF clock
Photoionisation of CS$_2$

S-$L_{2,3}$ resonant absorption

$$6\sigma_g^{-2}2\pi_g^{-1}/5\sigma_u^{-1}3\pi_g^{-1}/2\pi_g^{-1}2\pi_u^{-1}$$

Counts (arbitrary units)

Time of flight (ns)

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Photoionisation of CS$_2$ S-L$_{2,3}$ resonant absorption

![Graphs and diagrams showing binding energy and kinetic energy distributions for different ions and transitions.]

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Photoionisation of CS$_2$ S-L$_{2,3}$ resonant absorption

\[
\frac{6\sigma^{-1}2\pi^{-1}g^{-1}}{5\sigma^{-1}3\pi^{-1}u^{-1}/2\pi^{-1}2\pi^{-1}u^{-1}}
\]

\[
\frac{6\sigma^{-2}}{5\sigma^{-2}u^{-1}/6\sigma^{-1}5\pi^{-1}u^{-1}}
\]

Counts (arbitrary units)

Kinetic energy (eV)

Binding energy (eV)

(3h-1p)

valence ionization

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S-L$_{2,3}$ resonant absorption

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- Counts (arbitrary units)
- Kinetic energy (eV)

- 6$\sigma_g^{-1}$2$\pi_g^{-1}$ / 5$\sigma_u^{-1}$3$\pi_g^{-1}$ / 2$\pi_g^{-1}$2$\pi_u^{-1}$
- 6$\sigma_g^{-2}$ / 5$\sigma_u^{-2}$ / 6$\sigma_g^{-1}$5$\sigma_u^{-1}$
- Valence ionization
- (3h-1p)

- Time of flight Hit1 (ns)
- Time of flight Hit2 (ns)

- C$^+$
- 2$\pi_g^{-2}$
- S$^+$

- 3$\sigma_g^{-2}$ / 5$\sigma_u^{-2}$ / 2$\pi_g^{-1}$2$\pi_u^{-1}$
- 3$\sigma_g^{-1}$5$\sigma_u^{-1}$
- (3h-1p)

- CS$^+$
- S$^+$

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A three-body break-up is coplanar

Dalitz Plot: correlated momentum of the three fragments in terms of reduced energy

Visualize precursor ion geometry at the time of break-up
A three-body break-up is coplanar

Dalitz Plot: correlated momentum of the three fragments in terms of reduced energy

Visualize precursor ion geometry at the time of break-up
Photoionisation of CS$_2$

CS$_2^{2+}$ (3$h - 1p$) $\rightarrow$ C$^+$ + S$^+$ + S

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Branching ratios of dissociation channels are highly dependent on the transient molecular ion state.

Fragment ion kinematics clearly show effects of shell selective excitation, interpreted in terms of the energy sharing between the ejected electrons and molecular ion, and the subsequent conformal changes in the molecular ion.

These results bring us closer to being able to make a comparison between measured and computed differential cross-sections.
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*Review of Scientific Instruments, 84 073101 (2013)*

*Journal of Chemical Physics, 139 164309 (2013)*
What Next?

This Field

- Improve on the shell-selectivity – by better electron analysis and a smarter correlation strategy, higher photon energies, better energy resolution
- Orientation effects and angular distributions with shell selectivity to be explored
- Other perturbation regimes
  - Ion impact studies of similar type (MeV class ion accelerator: IUAC)
  - These effects in intense laser fields
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- Look into applications of ion–ion correlation mapping
  - Inspiration: stopping power studies from the 1960s has benefited isotope geochronology via accelerator mass spectrometry
  - Inspiration: atmospheric aerosol analysis has benefited from high resolution ToF-MS
What Next? Plans for IISER-Pune

- Teaching Plans: (re-)building classic experiments that form the bedrock of modern science, for interpretation by undergraduate students.
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Positron and Positronium collisions with molecules
What Next? Plans for IISER-Pune

- Teaching Plans: (re-)building classic experiments that form the bedrock of modern science, for interpretation by undergraduate students
- Positron and Positronium collisions with molecules
- Cross-disciplinary studies
▶ Development of laboratories for undergraduate teaching that foresee
  ▶ interesting and challenging experiments in normal courses
  ▶ Re-building classic experiments that form the bedrock of modern science, for
    interpretation by undergraduate students
  ▶ lead time of about two years

▶ In the long run:
  ▶ public outreach, attracting college teachers etc
  ▶ become a nodal centre supporting experimental activities for undergraduate
    Physics courses throughout the country, and bring a unique distinction to
    IISER Pune
Electrons and positrons scatter differently from atoms and molecules – theoretically well understood with fair experimental evidence in support.

Intriguingly, the positronium – a positron-electron bound state – has been found to scatter from molecules in a manner similar to electrons.

Positron and positronium sources are scarce – validation of experimental data is a hurdle; experiments continue to show interesting features.

Experimental facility is complex and lead time likely to be long.

Diagnostics used for electron collisions are readily applicable to positron collisions, and to a lesser extent to positronium collisions.

Only two or three facilities world-wide for such studies, none of them in India.
Experimental research at IISER Pune having a common knowledge base and technology
- plasmonics, nanophotonics, nanotechnology, magnetic systems, surface science, precision spectroscopy, quantum optics,
- gas phase laser spectroscopy and photoionization dynamics of biologically relevant molecules

Common thread: ever deeper understanding of the interaction of light with matter in a wide gamut of characteristics of the light
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Develop a long term programme initially focusing on laser–matter interactions in the strong field, short pulse regime
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Develop a long term programme initially focusing on laser–matter interactions in the strong field, short pulse regime

- A femto-second high power laser
- A pulsed noble gas cluster source for HHG
- A monochromator for the resulting VUV and XUV radiation pulses
- Opens up the possibility of time-resolved or ultrafast probing of molecules and condensed matter
- The monochromatic, short pulse length XUV radiation source would be useful for condensed matter studies, mainly for investigating electron correlations and magnetisation dynamics in real-time
- Can also be a source of pulsed high energy electrons with unusual properties – could be suitable for electron microscopy of surfaces and nanoparticles