



ELECTRON AND POSITRON INDUCED CHEMISTRY



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Book of Abstracts

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Elastic Electron Scattering with Molecules and Progress Report on Experiments with Biomolecules

Hyuck Cho, Chungnam National University, Physics Department

Absolute differential cross sections (DCS) for elastic electron scattering by molecules will be discussed, taking water vapour as an example. DCS's for water are measured for incident electron energies between 4 to 50 eV and over scattering angles of 10° to 180° using a crossed beam electron spectrometer. A magnetic angle-changing device based on the suggestion of Read and co-workers has been used to extend the measurements to backward angles (125° to 180°). The elastic integral and the momentum transfer cross sections are derived from these elastic differential cross sections.

More experimental studies of electron scattering with biomolecules are being prepared and the details will be reported.

One-Electron Resonances in Electron-Molecule Scattering

Robert R. Lucchese, Department of Chemistry, Texas A&M University

Resonant process can provide significant enhancements in the rate for a variety of process induced by electron collision with molecules. The simplest such resonance is a one-electron, or the shape resonance. We have developed an approach that can rapidly locate such resonances in larger molecular systems and can yield qualitative information about the location of the extra electron in the resulting temporary negative ion. Using this approach we have considered a number of molecular system from N₂ to C₆₀. Recent results on the resonances in electron scattering from formic acid, uracil, and glycine will be presented with a discussion on how those results can be used to help understand experimental dissociative attachment experimental data.

Cross sections for electron collisions with water molecules

Y. Itikawa, Institute of Space and Astronautical Science

Cross section data have been compiled from the literature for electron collisions with water molecules. [1] All major collision processes are reviewed: total scattering, elastic scattering, momentum transfer, excitation of rotational, vibrational, and electronic states, ionization, electron attachment, dissociation, and emission of radiation. In each case the collected data have been assessed and the recommended values of the cross section have been provided. In assessing the data, we have encountered several problems about the quality of them. In particular the following issues are discussed here:

- (1) A significant disagreement between the theoretical and experimental data on the elastic cross section in the low-energy region.
- (2) A lack of reliable data on the vibrational cross section near threshold and no clear evidence of resonance.
- (3) No beam-type measurements of the cross section for the excitation of electronic states.
- (4) Overall consistency of the available cross section data.

[1] Y. Itikawa and N. Mason, J. Phys. Chem. Ref. Data (submitted).

Some Chemical Applications of Electron Scattering

Josef Michl, Donald E. David, Duška Popović, and Dean Antic, Department of Chemistry and Biochemistry, University of Colorado, Boulder, CO 80309-0215, U.S.A.

The design, construction, and testing of an Electron Energy Loss Spectrometer (EELS) optimized for rapid measurement of vibrationally or electronically inelastic electron scattering by condensed thin films and dilute matrices of organic molecules at low temperatures will be described. The instrument is equipped with a multichannel energy analyzer that improves the detection efficiency by a factor of 50-100 relative to the usual single-channel detector. The films are formed on an atomically flat silver crystal cooled by a liquid helium cryostat. The high efficiency is critical to our application because of very low count rates for measuring electronic EELS due to small electron scattering cross sections. The instrument can be also used to measure inelastic loss spectra of gaseous molecules.

Two types of results will be presented: (i) solid-state electronic spectra of *endobenzotricyclo* [4.2.1.0_{2.5}]nonane (**1**), a polycyclic benzene derivative, and (ii) gas-phase vibrational spectra of propane as a function of incident energy and scattering angle. The results for **1** reveal a relatively rapid conversion into a product with a polycyclic *o*-xylylene structure as a result of an electron-impact induced electrocyclic transformation. The results for propane are compared with calculations done by P. Nárský and his collaborators in Prague, Czech Republic, using the Discrete Momentum Representation method, which he recently extended to vibrationally inelastic scattering of electrons by polyatomic molecules.

Comments will be made on the potential utility of electron scattering spectroscopy for the study of spin-forbidden and symmetry forbidden transitions in organic reactive intermediates.

Ionization energies of van der Waals clusters

Olof Echt, University of Innsbruck

We report on measurements of the appearance energies of van der Waals clusters by electron impact ionization. The results will be compared with published experimental and theoretical data.

Low-energy Electron Scattering from Closed-shell Diatomics

Slawomir Telega, Gdansk University of Technology, Faculty of Applied Physics and Mathematics

The quantum treatment of the collisional excitation of rotations and vibrations in closed-shell diatomic gases is carried out within a space-fixed frame of reference by solving the multichannel close-coupled equations. The interaction potentials contain the static, exchange and correlation-polarization contributions and are obtained via a local, multipolar expansion formulation from *ab initio* data. Numerical results are presented for H₂ and N₂ molecules.

Site selective electron attachment to the partially deuterated thymine

Sylwia Ptasińska, Leopold-Franzens-Universität Innsbruck, Institut für Ionenphysik, Technikerstr. 25, 6020 Innsbruck, Austria

The interaction of low energy electrons with compounds of DNA leading to negative ions gives insight into radiation damage processes in human cells. Our previous measurements carried out for nucleobases (e.g. thymine) demonstrated a very effectively decompose gas phase of these molecules by the electron attachment. In present studies we use the partially deuterated thymine in order to experimentally identify the site of selective electron attachment to this molecule. The resonance structures of negative ions of thymine formatting by the abstraction of radical or two hydrogen atoms compared with corresponding anions formatting by the abstraction of hydrogen (H) or/and deuterium (D) atoms from deuterated thymine and the formation H^- and D^- ions from (un)deuterated thymine allowed point out the position of the hydrogen atom in the molecule, where it is removed most efficiently.

The comparison D^- ions formation from the deuterated thymine with previously reported single and double strand breaks from the plasmid DNA shows that strand breaks are observed in the energy range where the hydrogen atom is abstracted from carbon sites.

This observation has significant consequences for understanding the molecular picture of radiation damage of living cells due to the secondary component of the high energy radiation.

Applications of the Feshbach-Fano-R-matrix method to electron-molecule scattering

Vincent Brems, Department of Chemical Physics, J. Heyrovsky Institute of Physical Chemistry, Dolejskova 3, 18223 Praha 8, Czech Republic

The Feshbach-Fano R-matrix (FFR) method has been lately developed [1] and successfully applied by Nestmann and co-workers to resonant electron-molecule collisions [1,2,3,4]. This method is based on the application of the Feshbach-Fano projector operator method within the R-matrix framework. The method allows one to project the background and resonant component out of the total cross section. This is not only usefull in order to rationalize fixed-nuclei R-matrix results but also in order to diabatize the R-matrix potential curves and perform nuclear dynamic computations.

This talk introduces the FFR theory and focuses on the applications. In particular the study of the low-energy resonant electron attachment to the fluorine and ozone molecules will be discussed in more details.

[1] B. M. Nestmann. J. Phys. B, 31 (1998) 3929

[2] T. Beyer, B. M. Nestmann and S. D. Peyerimhoff. J. Phys. B, 33 (2000) 4657

[3] T. Beyer, B. M. Nestmann and S. D. Peyerimhoff. J. Phys. B, 34 (2001) 3703

[4] V. Brems, T. Beyer, B. M. Nestmann, H.-D. Meyer and L. S. Cederbaum. J. Chem. Phys., 117 (2002) 10635

Sensitizing to Secondary Electron Damage

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Treatment of cancer represents one of the major challenges to medical research [1]. Radiotherapy, combining the use of ionizing radiation and the incorporation of cytotoxic agents within the tumour cells, has been developed as an alternative treatment to the traditional radical surgery, and 5-halouridines (e.g., 5-bromodeoxyuridine) have been suggested to be one of the most promising radiosensitizers for targeted therapies [2]. It has been shown that the substitution of thymidine nucleoside in the genetic sequence of cellular DNA by its halogenated analogs does not change the normal gene expression in non-irradiated cells, but increases sensitivity of the living cells [3] to X-rays by a factor of 4 [4]. It has been proposed that sensitization in the “halogen-modified” nucleic acids is presumably attributed to the interaction of hydrated electrons (e_{aq}^-) with the thymine surrogates [5]. However, not only such solvated electrons are capable to induce genotoxicity to cells but also the ballistic low-energy (< 20eV) electrons [6], which are the most predominant secondary species produced at the early time (fs-ps) after depositing energy into irradiated biological medium [7].

Here, we present measurements of Dissociative Electron Attachment (DEA) to the radiosensitizers (e.g., bromouracil, BrU [8], and bromouridin, BrdU [9]) in the gas phase. We will show, that at subexcitation energies (i.e., below the first electronic state of the molecule), electrons are able to effectively decompose BrU into Br^- and UyI^- (uracil-yl) and BrdU into Br^- and $BrUyI^-$ (bromouracil-yl). These results will be discussed within the context of recent theoretical works [10] and their application in radiotherapy.

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- [3] S. Zamenhof, R. DeGiovanni, S. Greer, *Nature* **181** 827 (1958)
- [4] W. Szybalski, *Cancer Chemother Reports* **58** 539 (1974)
- [5] M.D. Sevilla, R. Failor, G. Zorman, *J. Phys. Chem.* **78** 696 (1974)
- [6] L. Sanche, *Mass Spectrometry Reviews* **21** 349 (2002)
- [7] S.M. Pimblott, J.A. LaVerne, A. Mozumder, N.J.B. Green, *J. Phys. Chem.* **94** 488-495 (1990)
- [8] H. Abdoul-Carime, M.A. Huels, L. Sanche, E. Illenberger, *J. Am. Chem. Soc.* **123** 5354 (2001)
- [9] H. Abdoul-Carime et al. (submitted 2004)
- [10] X. Li, M.D. Sevilla, L. Sanche, *J. Am. Chem. Soc.* **125** 8916 (2003)

A Fluorescence-Based Method for Analyses of Low-Energy Electron Damage to DNA

Tihomir Solomun and Eugen Illenberger, Free University Berlin, Institute for Chemistry, Physical and Theoretical Chemistry, Takustrasse 3, D-14195 Berlin, Germany

Various active centres containing DNA molecules (single stranded oligonucleotides) were immobilized in an array on a gold surface and exposed to low-energy electron beam at different electron energies (<3eV) and doses. The electron damage was analysed by the binding ability of the complementary sequence as detected by a fluorescence method.

Recent Progress in the Development of the Discrete Momentum Representation Method for Electron Scattering by Polyatomic Molecules

Petr Carsky, J. Heyrovský Institute of Physical Chemistry, Academy of Sciences of the Czech Republic, Dolejškova 3, 182 23 Praha 8, Czech Republic

The purpose of this presentation is to show the recent progress in the development of the Discrete Momentum Representation Method (DMR) achieved by the Prague group. DMR is a rigorous ab initio method for elastic and vibrationally inelastic electron scattering by polyatomic molecules in the static exchange approximation. Results obtained so far for H₂, H₂O, CH₄ and C₃H₈ show that DMR can be a useful tool for interpretation of vibrational electron energy loss spectra of polyatomic molecules. Plans for the future work will be outlined: application to larger molecules, inclusion of polarization term into the interaction potential, and examination of the obtained results for a better understanding of the mechanism of vibrational excitation by electron impact.

Isotope effects in low-energy electron-molecule collisions

Jiri Horacek, Charles University, Prague, Czech Republic

Isotope effects in dissociative attachment and vibrational excitations will be discussed. Conditions for the appearance of the inverse isotope effect will be considered.

Calculations for vibrational excitations of methane by electron impact

Roman Curik, J. Heyrovsky Institute of Physical Chemistry, Prague

Differential and integral cross sections are presented for the vibrationally inelastic electron-methane scattering in the energy regime 1-30 eV. They were calculated by the Discrete momentum representation (DMR) approach. Two-channel Lippmann-Schwinger equation is solved in the momentum space. Interaction between scattered electron and methane molecule is described at exact static-exchange level. Nuclear motion of the molecule is assumed to satisfy harmonic approximation. Results are compared with the previous Single Centre Expansion calculations of Nishimura and Gianturco (2002) and available experimental data.

Electron impact ionization of alanine

Stefan Matejčík, Department of Plasmaphysics, Comenius University, Mlynska dolina F2, Bratislava, Slovakia

We will present the results of crossed electron molecule beams study of electron impact ionization reaction to the gas phase amino acid alanine (CH₃-CH(NH₂)-COOH). The mass spectra and mass-selective electron energy dependent ion yields have been measured. The high resolution gas phase ion energetic data have been estimated from the experimental data.

Positron and Electron scattering processes with polyatomic gases

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During the talk we will present a review of the second year of joint research within the EPIC European research Project in the theoretical and computational aspects of low energy scattering off polyatomic gases of both leptonic projectiles which are the probes employed in the network research activities, i.e.: electrons and positrons. In particular, it will be shown that some systems behave in similar, but yet different, ways when they undergo elastic and/or inelastic scattering by electrons and positron impact. Examples will be given for the case of CF_4 , a polyatomic target of interest in the world of nanofabrication of electronic chips, in relation of the conjectured existence of a Ramsauer-Townsend Minimum in the integral cross sections from both electron and positron scattering. Another example will be provided by the vibrational excitation efficiency of both CF_4 and H_2O , at low collision energies, by impact with electrons and positrons. In the cases where they exist, comparison will also be made with experimental findings. Finally, the importance of the formation of transient negative ions (TNI's) by secondary electron attachments to biologically relevant polyatomics will also be described and discussed.

Positron impact ionization of water

*Z. Pesič, Department of Physics and Astronomy, University College London, Gower Street, WC1E
6BT, UK*

The interaction of electrons and positrons with water molecules are extremely important for different areas, e.g. astrophysics, plasma physics etc. While different aspects of electron-water interaction have been studied extensively (e.g. [1] and references therein), very few studies have been reported concerning positron water interaction [2, 3].

Within the EPIC Network, the UCL positron physics group is investigating ionization and positronium formation cross-section of the water molecule. An overview of the UCL experimental techniques for measurements and of ionization cross section (direct and total) and for normalization will be presented [4, 5]. Progress in the measurement of water ionization cross section will be discussed and the first experimental data for the ionization cross section of water by positron impact will be presented.

- [1] G. Karwarsz et al, Riv. Nuovo Cimento 24 (2001)1
- [2] O. Sueoka et al, J. Phys. B: At. Mol. Opt. Phys 19 (1986) L373
- [3] F.A. Gianturco et al, PRA 64 (2001) 0327151
- [4] G. Laricchia et al, J. Phys. B: At. Mol. Opt. Phys. 35 (2002) 2525
- [5] P. Van Reeth et al, NIM B192, (2002) 220

Differential Studies of Ionization by Positron Impact

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Differential studies are important for understanding collision dynamics. An investigation is being carried out into the possible origins of a recently observed energy shift of $\sim 1.6\text{eV}$ between the theoretical and measured triply-differential cross-section for 50eV positron impact ionization of molecular hydrogen [1,2]. Two targets have been studied for the same final state energy. The final charge state of molecular deuterium was checked in order to establish possible dissociation contributions. A helium target was also employed to eliminate possible complications in the spectra arising from molecular excitations. Both the experimental data for D_2 and He have been found to be shifted as for the molecular hydrogen by $\sim 1.6\text{eV}$. Comparison with theories [3] and plans to extend these types of studies to the water molecules will be discussed.

[1] Á. Kövér, K. Paludan and G. Laricchia, J. Phys. B 34 L219, 2001.

[2] J. Fiol, V. D. Rodríguez and R. O. Barrachina, J. Phys. B 34 933, 2001.

[3] J. Fiol and Olson, J. Phys. B 35 1173, 2002.

Measurements of total cross sections for positron scattering in benzene

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Total cross sections for positron scattering on benzene at 1.5-20 eV have been measured with the new low-energy spectrometer in Trento [1]: it uses W thin film moderator, a longitudinal magnetic field (10 Ga), a 90° electrostatic bent; the scattering chamber is 10 cm long with 1 mm diameter apertures. Positron counting rate was 20-100 e^+/s - measurements are mean values of 20 measurements, 10 second each and over 5-6 runs. The statistical error is within 1%. The energy of the beam was calibrated against the threshold for positronium formation in N_2 (8.78eV).

Present data agree well with early measurements of Sueoka [2], but normalization of their data is needed by +19%. Our data at low energies disagree with newer results of Sueoka [3]; the agreement with theory is fairly good [4].

[1] Zecca A. et al., Rad. Phys. Chem. J., 68 (2003) 319-22

[2] Sueoka O., J. Phys. B 21 (1988) L631

[3] Kimura M. et al. Adv.Chem. Phys. 111 (2000) 537

[4] Gianturco et al., J.Phys. B 36 (2003) 1383

New data for positron scattering on molecules

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Italia*

Total cross sections for positron scattering on molecules (N_2 , C_6H_6 , $C_6H_5NH_2$, c - C_6H_{12}) from the new spectrometer in Trento [1] are presented.

In benzene (C_6H_6) the total cross section rises in the low energy limit, to high values of about 100 \AA^2 at 1 eV (30 \AA^2 is the cross section for electron scattering at the same energy [2]). In order to check if this high cross section is due to trapping of positron inside the benzene ring, the cross sections in aniline ($C_6H_5NH_2$) has been measured. We expected a smaller cross section due to the effect of localization of hybridized sp^2 electrons. Unexpectedly, the aniline cross section is identical to that in benzene, within our experimental error bar (less than 5%) in the entire 1-20 eV energy range. Both benzene and aniline cross sections show a "bump" slightly above the respective thresholds for positronium formation. No such bumps exist for positron cyclo-hexane scattering.

[1] Zecca A. et al., *Rad.Phys.Chem. J.*, 68 (2003) 319-22

[2] G.P. Karwasz, A. Zecca and R.S. Brusa, *Riv. Nuovo Cimento* 24 No.4, 1-101 (2001)

**The metastable decays of rare gas (Ar, Ne, Kr, Xe, and He) cluster ions:
experimental realization and theoretical treatment**

S. Denifl, J. Fedor, K. Gluch, S. Feil, S. Matt-Leubner, A. Stamatovic, F. Hagelberg, M. Probst, O. Echt, P. Scheier, T.D. Märk, Institut für Ionenphysik, Leopold Franzens Universität, Technikerstrasse 25, A-6020 Innsbruck, Austria

Metastable dissociation of rare gas cluster ions (He, Ne, Ar, Kr, and Xe) has been investigated experimentally utilizing the mass analysed ion kinetic energy (MIKE) technique with a three sector field mass spectrometer. Based on quantum chemical calculations we constructed theoretical MIKE peaks and compared them with the experimental data. By comparison with the experimental data we obtained information about the temperature of the neutral precursor. Excellent agreement between the experimental data and the theoretical model has been observed [1] depending on the potential energy curves used. In contrast to the Ar, Ne, Xe and Kr dimers we did not find a unimolecular fragmentation channel for He_2^+ . For the larger He_n^+ ($n=3\dots30$) cluster ions we observed the unimolecular loss of monomers driven by vibrational predissociation.

This work was partially supported by the FWF, ÖNB and ÖAW, Wien, Austria and the European Commission, Brussels (network programme).

[1] J. Fedor, R. Parajuli, S. Matt-Leubner, O. Echt c, F. Hagelberg, K. Gluch, M. Probst, P. Scheier, T. D. Märk, *Phys. Rev. Lett.* 91 (2003) 133401.

Low energy electron scattering from halogenated hydrocarbons

Nykola Jones, Dept. of Physics and Astronomy, University of Aarhus, DK-8000 Aarhus C, Denmark

A major theme of the EPIC network is to investigate the electron scattering properties of halogenated species of importance in the plasma etching industry. Data have important implications for the cooling and heating of electrons in plasmas in general. Cross-sections for elastic and inelastic scattering of electrons below a few hundred meV are important input not only for models of industrial plasmas, but also atmospheric and interstellar plasmas. Scattering data may also be used to show that species may undergo dissociative attachment at very low energy with very high cross-sections, for example exceeding 1000 \AA^2 .

Following the EPIC programme, we have explored the scattering properties and measured cross-sections for a number of halogenated hydrocarbons CH_xR_y and fluorocarbons CF_xR_y , where $\text{R} = \text{F}, \text{Cl}, \text{Br}, \text{I}$. In this presentation data will be reported for CF_4 , CCl_4 , CF_3I , CF_3Br , CH_3I , CH_3Cl , CH_3Br , CHCl_3 and for the atmospherically interesting molecule SF_5CF_3 . We have developed a means of obtaining absolute scattering cross-sections for dissociative attachment independently of any other data save our absolute scattering data. This method is at present limited to non-polar molecules, and cross-sections will be presented for Cl^- production from CCl_4 down to sub-thermal energies.

The Anomalous Shape of the Cross Section for the Formation of SF_3^+ Fragment Ions Produced by Electron Impact on SF_6

S. Feil, K. Gluch, P. Scheier, A. Stamatovic, K. Becker, and T.D. Märk, Institut für Ionenphysik, Leopold Franzens Universität, Technikerstrasse 25, A-6020 Innsbruck, AUSTRIA

The partial ionization cross section for the formation of SF_3^+ fragment ions following electron impact on SF_6 is known to have a pronounced structure in the cross section curve slightly above 40 eV. We used the mass-analyzed ion kinetic energy (MIKE) scan technique to demonstrate the presence of a channel contributing to the SF_3^+ partial ionization cross section that we attribute to the Coulomb explosion of doubly charged metastable SF_4^{2+} ions into two singly charged ions SF_3^+ and F^+ , with a threshold energy of about 45.5 eV. Thus the observed unusual shape of the SF_3^+ partial ionization cross section is the result of 2 contributions, (i) the direct formation of SF_3^+ fragment ions via dissociative ionization of SF_6 with a threshold energy of 22 eV and (ii) the Coulomb explosion of metastable SF_4^{2+} ions with a threshold energy of about 45.5 eV. A detailed analysis of the MIKE spectrum reveals an average kinetic energy release of about 5 eV in the Coulomb explosion of the SF_4^{2+} ions with evidence of a second channel corresponding to an average kinetic energy release of about 1.1 eV.

Imaging DNA damage by AFM

D. Mayr, Department of Physics and Astronomy, The Open University, Milton Keynes, MK7 6AA

Atomic force microscopy (AFM) can be used to image biological samples at nanometre resolution and therefore is well suited for the study of DNA damage. I will explain how the AFM works, show some results and speak about experiments planned for the near future.

Low-energy electron induced break-up of water

Chiara Piccarreta, University College London

Electron collisions with water are very important for the understanding of radiation damage in biological system, atmospheric processes, masers, etc... These collisions can result in the rotational, vibrational and/or electronic excitation of the molecule but they can also induce fragmentation. Specifically, low-energy electrons can produce fragmentation via dissociative electron attachment (DEA) and electronic excitation into dissociative states. The R-matrix method and the R-matrix polyatomic code is used to treat the electronic part of the process. We have performed fixed-nuclei calculations for 630 geometries of water. These are defined by 6 different angles and 14 different OH internuclear distances. Previous calculations and experiments show the presence of three Feshbach resonances below the ionisation threshold. Since the first four excited states of water are dissociative, excitation into any of these will produce fragmentation. Our model incorporates nine target states in the close-coupling expansion. The energies obtained for the ground state and the lowest singlet excited states compare well with more accurate calculations. Using this model, cross sections for electronic excitation into the first 4 excited states as a function of geometry were calculated. We will present resonance positions and widths as a function of geometry. These results will form the starting point for a full-dimensional study of the DEA and dissociative excitation processes.

Electron impact calculation on biological molecules

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00185 Rome, Italy*

Recent work on the scattering of electrons around 9eV on the uracil molecule will be discussed in detail. The behaviour of the eigenphase sums and of the resonant component of the scattering cross section will be analysed in order to confirm the presence of a dominant shape resonance. The effect of molecular bond stretching on the detachment of H⁻ from Uracil samples will also be analysed and discussed.

Spatial differential cross section maps for vibrational excitation by electron impact

Barbara Pezler, J. Heyrovsky Institute of Physical Chemistry, Czech Republic

There has been significant increase of activities in development and applications of ab-initio computational methods for the vibrationally inelastic scattering of electrons by polyatomic molecules. Despite of the recent work in this field, the mechanism of the vibrational excitation by electron impact is far from being understood. Therefore the main purpose of the present project is the study of spatial differential cross-section maps for the vibrational excitation of molecules using discrete momentum representation method. The energy range of our interest is 1-30 eV.

Theory of Vibrationally Inelastic Electron Transport through Molecular Bridges

Martin Cizek, Institute of Theoretical Physics, Charles University, Prague, Czech Republic

Vibrationally inelastic electron transport through a molecular bridge that is connected to two leads is investigated. The study is based on a generic model of vibrational excitation in resonant transmission of electrons through a molecular junction. Employing methods from electron-molecule scattering theory, the transmittance through the molecular bridge can be evaluated numerically exactly. The current through the junction is obtained approximately using a Landauer-type formula. Considering different parameter regimes, which include both the case of a molecular bridge that is weakly coupled to the leads, resulting in narrow resonance structures, and the opposite case of a broad resonance caused by strong interaction with the leads, we investigate the characteristic effects of coherent and dissipative vibrational motion on the electron transport. Furthermore, the validity of widely used approximations such as the wide-band approximation and the restriction to elastic transport mechanisms is investigated in some detail.

Calculations of the cross sections for vibrational EELS of cyclopropane

Michal Kovacic, J. Heyrovsky Institute of Physical Chemistry, Prague, Czech Republic

Ab initio Discrete Momentum Representation (DMR) method is used to calculate differential and integral cross sections for vibrational excitation of cyclopropane by electron impact. Three-dimensional Lippmann-Schwinger equation is solved by two-channel approach and harmonic approximation is taken to describe nuclear motion of the molecule. Static and exchange contributions to electron-cyclopropane interaction are evaluated explicitly. Results for the energy range 1-30 eV are presented and compared to available experimental data.

Cold electrons: virtual states and attachment

David Field, Department of Physics and Astronomy, University of Aarhus

The aim of my talk is to attempt to give insight into the physical processes which may occur when cold electrons encounter non-polar molecules. Recent experiments involve collisions of electrons with a variety of molecules such as benzene, CO₂, C₆F₆, para-xylene, para-difluorobenzene with collision energies down to a few meV. Results show that scattering cross-sections can achieve very large values of several hundred to in excess of one thousand Å² at very low energies. I will discuss the physical basis for this phenomenon, called "virtual state scattering". I will also discuss how virtual state scattering may act as a gateway to temporary negative ion formation. This has interesting implications for negative ion formation in industrial plasmas and also in natural plasmas such as the upper atmosphere of the Earth and the interstellar medium.

Theory of resonances: determination of energies, widths and cross sections by the Feshbach-Fano R-matrix method

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The study of inelastic resonant electron-molecule collisions splits into two parts. The electron scattering by the molecule with its nuclei fixed is investigated and then the nuclear dynamics is considered using the results of the fixed-nuclei calculations as the input. The two parts of the problem are treated with different methods. Well developed and widely used approach of the study of the fixed-nuclei problem is the R-matrix theory. For the nuclear dynamics the most suitable treatment is the Feshbach-Fano projection-operator approach. Therefore it is very desirable to join these two theories to obtain a unified method of solving the whole problem.

The Feshbach-Fano R-matrix (FFR) method has been developed lately and applied successfully by Nestmann and co-workers to resonant molecule collisions, see e.g. [1]. The method allows one to project the background and resonant component out of the total cross section and determine the resonance parameters. In order to establish the FFR method as a general scheme for treating resonant scattering we have studied it on model problems.

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Quantum Wave Packet Dynamics: Electron Induced Dissociation of Water

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A time-dependent description of the dissociative attachment and impact dissociation of water excited by an impinging electron is proposed. Water is the key molecule for all known life forms, yet how it fragments in collisions with low-energy electrons remains uncertain. Theoretically this problem involves treating the interplay between the electron scattering dynamics and the nuclear motion dynamics. No previous study of dissociative attachment has considered more than a single nuclear motion coordinate but the different possible dissociation products for water make such a full three-dimensional nuclear motion study essential. Electron scattering will be treated using the fixed nuclei approximation and the R-matrix codes. Calculations at many internuclear configurations will give surfaces for the three low-lying, long-lived resonance states whose excitation leads to either dissociative attachment or autoionisation, which is often accompanied by vibrational excitation of the target. Wavepacket methods are being used to treat the nuclear motion. A new code based on wavepacket method is being developed to treat the nuclear motion. The results we are going to obtain will be of importance not only for radiation damage in biological systems but also regimes where electron collisions with water are important such as the physics of the upper atmosphere and astrophysics.

Interaction of Activated Oxygen and Water with Bare and Hydrogenated Diamond Surfaces

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Hydrogen-terminated diamond films have been considered as possible candidates for various electronic and optical applications due to their surface conductivity, chemical inertness, and negative electron affinity (NEA), which results in the enhancement of electron emission when irradiated with electrons or photons. However, even brief exposure of hydrogenated diamonds to air results in gradual deterioration of their electron emission properties and shifts the EA from negative to positive. This decay was found to be correlated with surface oxygen, the origin and adsorption mode of which were speculated on but unidentified. Possible oxygen-containing reactants may include molecular oxygen, water vapor, etc.

It is the aim of the present work to study the chemical state of hydrogenated and bare polycrystalline diamond films in situ exposed to water vapor and to oxygen. This task is achieved by X-ray photoelectron spectroscopy (XPS) utilized for quantitative elemental analysis of the near-surface region, high resolution electron energy loss spectroscopy (HREELS) and ESD measurements by which the surface bonding configuration was established

Electron Attachment to molecules of biological relevance

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We have studied electron attachment to a series of DNA/RNA bases using a crossed electron/neutral beams technique in combination with a quadrupole mass spectrometer. In several cases electrons at energies below the threshold for electronic excitation can lead to decomposition. Gas phase uracil decays into a mobile hydrogen radical and the corresponding closed shell uracil fragment anion (U-H)⁻. We have determined absolute dissociative attachment cross sections for such hydrogen loss reactions which can reach up to approximately $1.2 \times 10^{-19} \text{ m}^2$ for thymine, $2.3 \times 10^{-20} \text{ m}^2$ for cytosine and $5 \times 10^{-20} \text{ m}^2$ for glycine (G-H)⁻. In addition we report on our studies regarding the formation of anions from desoxyribose sugar. In this case the dehydrogenation channel is not the dominant reaction channel. Instead, the anion with the highest dissociative electron attachment cross section of desoxyribose is formed by the release of neutral particles equal to two water molecules. Such results have important bearing regarding the question of exposure of living tissue to ionizing radiation. Energy deposition into irradiated cells produces electrons as the dominant secondary species. After irradiation these electrons exist as ballistic electrons with an initial energy distribution up to several tens of electron volts.

Complete chemical transformation of a molecular film by subexcitation electrons (< 3 eV)

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The potential of slow electrons to act as a soft tool to control a chemical reaction in the condensed phase is demonstrated. By setting the energy of a well defined electron beam to values below 3 eV, the surface of a thin film of 1,2-C₂F₄Cl₂ molecules can completely be transformed into molecular chlorine (and byproducts, possibly perfluorinated polymers). At higher energies (above the threshold for electronic excitation, > 6 eV) some equilibrium composition between product and initial molecule can be achieved, however, accompanied by a gradual overall degradation of the film. The effect of complete transformation is based on both the selectivity and particular energy dependence of the initial step of the reaction which is dissociative electron attachment (DEA) to C₂F₄Cl₂ [1-3] but also the fact that the initial molecule is efficiently decomposed by subexcitation electrons while the product Cl₂ is virtually unaffected. This is due to the fact that s wave electron attachment is symmetry forbidden to ground state Cl₂ [4]. The progress of the reaction during electron irradiation is followed by electron stimulated desorption of Cl⁻ as a probe for the concentration of Cl₂ and C₂F₄Cl₂ at the surface of the irradiated area.

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The formation of the amino acid glycine in electron irradiated extraterrestrial ices

Philip Holtom, University College London

The interstellar medium - the vast space between the stars - is a rich reservoir of molecular material ranging from simple diatomics to more complex, astrobiologically important molecules such as cyanopolyacetylenes and the carbon hydrate glycolaldehyde. During the last years, observational astronomy has refined its spectroscopic detection methods, and particular attention has been devoted to identifying interstellar bio-molecules that may provide important insights into the history of the solar system and the origins of life on Earth. We will present the results of recent studies on the formation of the bio-molecule glycine in an electron irradiated interstellar ice analogue composed of Methylamine (CH₃NH₂) and Carbon Dioxide (CO₂) ice. We will discuss the formation and detection of glycine, and its relevance to origins of life on earth and discuss possible formation mechanisms in the interstellar medium.

Preparation, characterisation and scattering of low energy electrons by hydrogenated silicon surfaces

D. Caceres, Université Paris Sud

Hydrogen-terminated diamond films have been considered as possible candidates for various electronic and optical applications due to their surface conductivity, chemical inertness, and negative electron affinity (NEA), which results in the enhancement of electron emission when irradiated with electrons or photons. However, even brief exposure of hydrogenated diamonds to air results in gradual deterioration of their electron emission properties and shifts the EA from negative to positive. This decay was found to be correlated with surface oxygen, the origin and adsorption mode of which were speculated on but unidentified. Possible oxygen-containing reactants may include molecular oxygen, water vapor, carbon oxides, etc.

In neither of those studies, however, the effect of water exposure on the bonding configuration of the diamond surface was considered. This makes the validity of different surface conductivity and photoemission theories questionable. Determination of such an effect is of great importance both for basic understanding of chemical processes on diamond surfaces at the conditions close to ambient, and for development of electronic and electro-optical devices based on diamond films.

It is the aim of the present work to study the chemical state of hydrogenated and bare polycrystalline diamond films in situ exposed to water vapor. This task is achieved by X-ray photoelectron spectroscopy (XPS) utilized for quantitative elemental analysis of the near-surface region, and high resolution electron energy loss spectroscopy (HREELS) measurements by which the surface bonding configuration was established.

In summary, water vapor molecules adsorb on both hydrogenated and bare diamond surfaces. Based on the analysis of the corresponding HREELS spectra, the interaction of water (a mixture of D₂O and H₂O) molecules with the bare surface results in dissociation of considerable amount of them and formation of C=O, C–O–D (C–O–H), and C–D (C–H) surface bonds. These chemisorbed structures are mostly stable toward 300°C annealing. Hydrogen-terminated diamond does not chemically react with water at the conditions applied in this work, but nevertheless, some physisorption of water on this surface takes place as observed by both XPS and HREELS

In summary, oxygen adsorption on the bare diamond surface results in formation of C–O–C and CO bonds, as well as molecularlike adsorbed CO. We resolved the uncertainty related to the location of the principal vibrational loss feature associated with C–O–C: it is characterized by the peak at 125 meV. The peaks at 220 and 270 meV are ascribed to CO and adsorbed "molecularlike" CO, respectively. The interactions of low energy electrons with the oxygen-exposed hydrogenated diamond surfaces have a more complicated nature. For the fully hydrogenated surface produced by ex situ MW hydrogen plasma treatment, the energy losses of the 4.5 eV primary electrons are mainly due to C–C, C–H, CO, O–H, and C–O– surface vibrations. In situ hydrogenation does not produce fully hydrogen-terminated surface, and exposure of this surface to thermally activated oxygen results in the formation of structures characteristic to both bare and fully hydrogenated diamond. The HREEL spectra alone do not permit, however, to establish the exact bonding configuration of chemisorbed oxygen. This task is achieved by correlation with the previously performed ESD measurements which rule out formation of C–O–C thus leaving CO and C–O–H as the only possible bonding configurations of oxygen on the hydrogen-terminated diamond surface.

Electron Induced chemistry; Future perspectives

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The ability to understand, manipulate and control physico-chemical processes at the molecular level is one of the great challenges of modern research and underpins the development of vibrant new technologies of the 21st century, for example the development of nanolithography. Such molecular engineering requires selective bond cleavage in target molecules to allow subsequent management of the local site chemistry. Recent research (including much in the present EPIC Network) has revealed that it is possible to influence the excitation and dissociation of molecules through the manipulation of electron interactions at the molecular level. Since electrons are ubiquitous in nature and electron induced reactions (in the gaseous phase, on surfaces and in the condensed phase) initiate and drive the basic physical-chemical processes in many areas of science and technology from industrial plasmas to living tissues our ability to control electron interactions provides exciting new opportunities that can now be exploited by both the research and technological communities.

In addition we now have the opportunity to utilise new instrumentation to study single molecular systems. For example, the development of the Scanning Tunnel Microscope (STM) -an electron emitter- has introduced the capability of atomic-scale imaging, analysis and individual atomic/molecular manipulation providing a new method for investigating single electron –molecule/atom interactions on a well defined spatial scale.

Furthermore the development of ultracold atom sources for collisional studies, the preparation of the first ultracold molecular targets and the formation of ultracold plasmas and atomic lattices will allow new electron collision phenomena to be investigated at ultracold temperatures, in turn initiating new forms of low temperature chemistry in which electron tunnelling may play a key role.

Thus the whole field of electron interactions with atoms/molecules and surfaces is currently enjoying a renaissance. Currently several European groups are at the forefront of such pioneering research but, in contrast to the USA and Japan, the European research effort has been fragmented and co-ordination poor. EPIC has, in part, addressed these issues bringing together Europe's leading experimental and theoretical groups in a large-scale, multidisciplinary and collaborative research programme that will both maintain its international excellence and establish Europe as a leading centre for investigation of molecular control through electron processing. Such research will have direct relevance in many areas from the basic sciences, through astronomy and radiation chemistry to industrial applications. In this presentation I will therefore look forward and review the new opportunities for the field. I will also discuss how the current EPIC Network may be developed and enhanced to take advantage of the new opportunities.

Posters

Cold Electron Scattering By A Chiral Molecule

Ingo Struve(1), Patrick Cahillane(2), Nykola Jones(1), Søren Vrønning Hoffmann(3), Lars Bojer-Madsen(1), David Field(1) and Jean-Pierre Ziesel(4)

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Using the ASTRID storage ring, we have investigated the interaction of electrons [1-4] with the D- and L-enantiomers of the chiral molecule butan-2-ol down to energies of a few meV. Cross-sections will be presented for integral scattering and for scattering in the presence of a 20 G axial magnetic field, which yields backward scattering cross-sections. Integral cross-sections for the D and L species show identical values within experimental error. Cross-sections measured in the presence of a magnetic field however appear to show significant differences, which are most marked in the tens of meV electron impact range.

These results may have interesting implications for the interaction of electrons with biological molecules and in particular for the formation of non-racemic mixtures of amino acids in space (see for example [5]).

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Low energy electron scattering from halogenated hydrocarbons

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A major theme of the EPIC network is to investigate the electron scattering properties of halogenated species of importance in the plasma etching industry. Data have important implications for the cooling and heating of electrons in plasmas in general. Cross-sections for elastic and inelastic scattering of electrons below a few hundred meV are important input not only for models of industrial plasmas, but also atmospheric and interstellar plasmas. Scattering data may also be used to show that species may undergo dissociative attachment at very low energy with very high cross-sections, for example exceeding 1000 \AA^2 .

Following the EPIC programme, we have explored the scattering properties and measured cross-sections for a number of halogenated hydrocarbons CH_xR_y and fluorocarbons CF_xR_y , where $\text{R} = \text{F}, \text{Cl}, \text{Br}, \text{I}$. In this presentation data will be reported for CF_4 , CCl_4 , CF_3I , CF_3Br , CH_3I , CH_3Cl , CH_3Br , CHCl_3 and for the atmospherically interesting molecule SF_5CF_3 . We have developed a means of obtaining absolute scattering cross-sections for dissociative attachment independently of any other data save our absolute scattering data. This method is at present limited to non-polar molecules, and cross-sections will be presented for Cl⁻ production from CCl_4 down to sub-thermal energies.

The Determination of Absolute Electron Attachment Cross-Sections from Beam Scattering Data

David Field(1), Jean-Pierre Ziesel(2), Nykola Jones(1), Søren Vrønning Hoffmann(3), and Lars Bojer-Madsen(1)

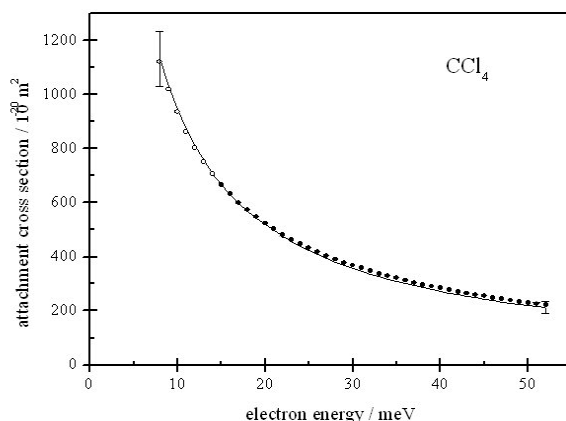
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The attachment of electrons to molecules is a very efficient means of producing negative ions and instigating chemistry. The process is important in the chemistry of halogenated hydrocarbon reactive plasmas, used in etching and deposition in device fabrication, and in other contexts such as the destruction of halogen-containing species in the atmosphere of the Earth. All cross-sections reported in the literature depend for absolute calibration on bulk rate constant measurements reviewed in [1]. We report here the first determination of absolute attachment cross-sections independently of rate constant values. This method depends on the measurement of absolute integral scattering cross-sections and cross-sections in the presence of an axial magnetic field (20 G) in the energy range of a few meV to 50 meV [2-7].

Theoretical analysis [2] shows the relationship between the coupled events of elastic scattering and attachment processes. Using a fitting procedure, we have used this analysis to extract absolute cross-sections for attachment of electrons to CCl_4 (to form Cl^-) and SF_6 . Results for CCl_4 (fitted and open circles) are shown in the figure. Data from [8], calibrated with rate constant data, is also shown as a solid line, showing strikingly good agreement with our derived values in this case. Similar, but rather less precise agreement is found for SF_6 [3].



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