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## Roadmap on photonic, electronic and atomic collision physics: II. Electron and antimatter interactions

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## Roadmap

# Roadmap on photonic, electronic and atomic collision physics: II. Electron and antimatter interactions

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## Abstract

We publish three Roadmaps on photonic, electronic and atomic collision physics in order to celebrate the 60th anniversary of the ICPEAC conference. In Roadmap II we focus on electron and antimatter interactions. Modern theoretical and experimental approaches provide detailed insight into the many body quantum dynamics of leptonic collisions with targets of varying complexity ranging from neutral and charged atoms to large biomolecules and clusters. These developments have been driven by technological progress and by the needs of adjacent areas of science such as astrophysics, plasma physics and radiation biophysics. This Roadmap aims at looking back along the road, explaining the evolution of the field, and looking forward, collecting contributions from eighteen leading groups from the field.

Keywords: collision physics, leptons, antimatter

(Some figures may appear in colour only in the online journal)

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## INTRODUCTION

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To celebrate the 60th anniversary of the ICPEAC conference, we publish a series of three Roadmaps on photonic, electronic and atomic collision physics. One for each of the three classes of projectile that comprise the breadth of topics encompassed by ICPEAC; *I. Light-matter interaction*; *II. Electron and antimatter interactions*; and *III. Heavy particles: with zero to relativistic speeds*. Each of the Roadmaps is intended to provide an overview of the present status of the field, how it was arrived at and address current and future challenges faced by those working in the broad area of research. As with all IOP Roadmaps, the three articles have been authored collaboratively by leading researchers in the areas and each aims to provide an impression of current trends in the respective field of research.

The field of research covered by this second Roadmap is at the heart of ICPEAC. The Roadmap provides glimpses into the future of the field by explaining important and promising theoretical and experimental trends and developments. It comprises eighteen contributions by leading scientists distributed over three topic sections: topic section 1 on electron scattering, topic section 2 on positrons and antimatter and topic section 3 on applications.

Due to continuous development of the computational and instrumental techniques and partly stimulated by the demands from neighbouring areas of science and technology the thriving field of electron scattering, dealt with in topic section 1, has considerably evolved over the years: Bray considers *ab initio* quantum methods which have succeeded in routinely providing accurate scattering cross sections for few-electron atomic targets and for simple molecules and which are currently developing into tools for studying more complex systems of interest to science and industry. Additional computational challenges arise when polarization, alignment and orientation are studied, in particular, on short time-scales. The contribution of Bartschat deals with this aspect of electron scattering. Murray discusses experiments on electron collisions involving laser-aligned targets which drive technical developments in laser stability and laser control and vice versa. Tennyson considers the increasing demands for reliable electron–molecule collision cross-sections made by applications such as low-temperature plasma chemistry or radiation biology which have led to the development of a powerful B-spline theoretical method which allows for a largely still to be implemented uncertainty quantification. Dorn describes novel kinematically complete electron-collision experiments which have been enabled by the seminal reaction microscope and which will greatly

benefit from ongoing developments of particle detectors and target sources. Yamazaki and Takahashi discuss time-resolved momentum spectroscopy which provides possibilities for 3D molecular wave-function mapping, viewing the stereo-dynamics of molecular ionization, and for monitoring chemical reactions. Whilst another experimental approach, that of dissociative electron attachment which even holds the promise to enable chemical control, is considered by Mason. Novotný and Wolf describe a new cryogenic storage ring in combination with an ultracold electron target that permits studies of collisions between electrons and molecular ions in their rotational and vibrational ground states, the very conditions which prevail in the interstellar medium. Electron-collision studies with biomolecules have revealed that secondary low-energy electrons, produced by ionizing radiation in living tissue are predominantly responsible for the ensuing radiobiological damage. Sanche describes ongoing experimental and theoretical developments, of target preparation and of Monte-Carlo codes, respectively, aimed at obtaining a complete knowledge of electron induced damage of living cells. Centurion describes ultrafast electron diffraction which has been proven to be an excellent tool for following the nuclear motion in molecules induced by ultrafast laser pulses. The challenge to improve spatial and temporal resolution of this intriguing experimental technique will be taken up by devising and applying pulsed photo-electron guns with megahertz repetition rates.

The second topic section of this Roadmap deals with antimatter which has fascinated both scientists and the general public since its discovery, shortly after its existence was predicted by Dirac 90 years ago. One of the big scientific questions related to antimatter is why antimatter and matter do not occur in equal amounts in the observable, at least for us, part of the universe. Another intriguing question is how gravitation acts on antimatter. Tackling these questions experimentally requires the availability of neutral antimatter. Yamazaki considers schemes that have been devised to produce antihydrogen ( $\bar{\text{H}}$ , the simplest antiatom) and other neutral antimatter by ‘atomic’ collisions involving, in the case of  $\bar{\text{H}}$ , antiprotons and positrons. Current experimental efforts aim at increasing the efficiency of production processes by using advanced trapping and particle-cooling techniques. Antiprotons for these experiments are at present only available at CERN. Laricchia considers positrons, which in contrast can be obtained from laboratory sources, and consequently their use is much more widespread. Scattering experiments with positrons and positronium offer unique possibilities for studying unusual aspects of electromagnetic interactions in small quantum systems. Surko discusses positron traps which allow for accumulation and for cooling and compression of the trapped charge cloud, with one of the goals being the creation of a positronium Bose–Einstein condensate, which could also serve as a source for short-pulsed positron beams of unsurpassed brilliance. Sullivan considers the challenges that need to be addressed to obtain a thorough understanding of positronium formation and other antimatter related phenomena in positron scattering from atoms or molecules which is still lacking. On the experimental side significant

improvements are expected from ongoing instrumental developments, such as techniques for the preparation of dense targets of metal atoms and (bio)molecules and, ultimately, a positron reaction microscope. Likewise, an extension of the theoretical methods to more complex targets is required for ultimately understanding the interactions of positrons on cosmic scales (including interactions with dark matter) and in technical applications such as positron emission tomography for medical diagnostics and positron spectroscopy in the materials sciences, these aspects are described by Gribakin.

Applications which have been the driving force also of much electron-collision work are considered in topic section 3. Savin considers the requirement for a huge amount of atomic and molecular collision data, of ever increasing accuracy, to facilitate the unravelling of the physics of cosmic plasmas. This requirement is driven by the increasing sensitivity and sophistication of astronomical instruments and is a challenge that is being met on the experimental side by new developments in particle generation and storage technology. Y Ralchenko considers our ability to accurately analyse and model hot magnetically-confined fusion plasmas, which are

governed by electron-ion collisions, which is an important asset in the global effort to provide a clean energy source for future generations. Leptonic collisions are also important in the hot dense, laser-generated plasmas which serve as light sources for EUV lithography in the semiconductor industry. Hoekstra and O'Sullivan consider this application where (cost-)efficiency is a particular demand spurring intense applied research.

We hope that our readers will share our opinion that the contributions to this Roadmap, introduced above, which are representative of many more related scientific activities, bear vivid witness that electron and antimatter collisions are thriving now as ever during the past sixty years of ICPEAC and still hold a large discovery potential in fundamental and applied research.

### Acknowledgments

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## ELECTRON SCATTERING

### 1. Electron collision theory

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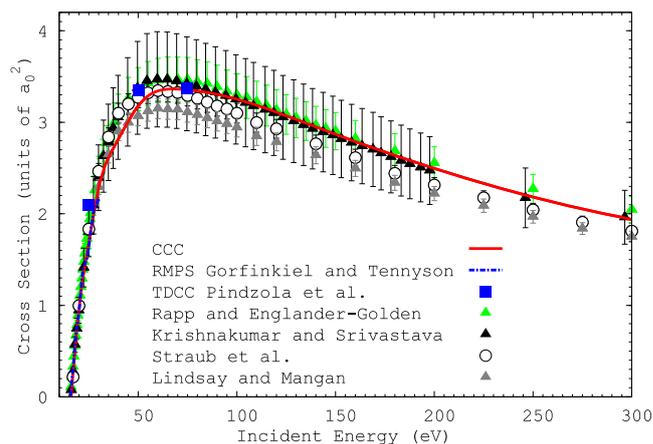
**Introduction.** The field of electron collisions in atomic and molecular physics is quite old, with the foundations for modern-day approaches laid down in the early 1930s by Massey and Mohr [1]. The problem complexity has several aspects:

1. The total wavefunction has to have the appropriate symmetry due to electrons being identical fermions.
2. The targets have countably infinite discrete bound (negative-energy) states.
3. The targets have an uncountably infinite continuum of free (positive-energy) states.
4. Upon ionization three free charged particles interact via the long-ranged Coulomb potential out to infinite separation.

The first three points above are of a computational nature, but the last makes the ionization problem particularly difficult to even formulate. Yet, through unitarity ionization is connected to all other collision processes, and thereby questioning the validity of results for any collision process above the ionization threshold.

**Theoretical progress.** The computational complexity of the problem requires considerable computational power, which became available in the 1990s. Extensive computational methods evolved following the close-coupling formalism of Massey and Mohr [1], while ignoring explicit treatment of ionization. So it was a considerable surprise when the convergent close-coupling (CCC) method was able to reproduce the measurements of the e-H total ionization cross section and spin asymmetry by associating ionization with excitation of positive-energy pseudostates [2]. This showed that the first three points above could be solved computationally. To check the importance of the last point the CCC formalism was applied directly to fully differential ionization processes, again with considerable success [3]. Only several years later the Coulomb problem was formulated correctly [4], and the explanation for the success of existing computational methods has been provided [5]. The resulting conclusion, for sufficiently simple targets, is that the close-coupling formalism is able to fully solve the electron scattering problem irrespective of energy or the scattering process of interest.

**Motivation.** Given that all four aforementioned points have now been effectively solved, even if only for sufficiently simple targets, it is important to consider what the motivation is for continued development of the field. This is now to



**Figure 1.** e-H<sub>2</sub> total ionization, see [7] for details. Reprinted with permission, copyright 2016 by the American Physical Society.

provide accurate electron collision data for atoms and molecules of interest to science and industry.

Collisions on the atomic scale go on all around us, and typically involve targets much more complicated than atomic hydrogen. So complexity in the general electron-atom collision problem arises from the number of active electrons involved. For molecular targets there are also new degrees of freedom of rotation and vibration. How to manage the complexity of general collision systems that may be of practical interest is not a solved problem.

**Atomic targets.** Quasi one- and two-electron atoms and ions, where the interest is in the interaction with the valence electrons is mostly computationally manageable, see for example electron-impact ionization with excitation of helium [6]. However, when it comes to atomic targets such as tungsten, of importance to the fusion reactor ITER, then the problem of treating all of the valence electrons systematically to convergence is not yet feasible. So one area of further development is the treatment of multi-electron atoms, of importance to science or industry, in such a way so as to perform scattering calculations that systematically check the treatment of all of the interacting electrons.

**Molecular targets.** Electron scattering on molecular targets is today where electron-atom scattering was two decades ago. We can now confidently say that electronic excitation and ionization of the H<sub>2</sub> molecule can be calculated as accurately as that for H, see figure 1 for example.

A most satisfying test of theory is when calculations yield results that are contrary to existing experiments, and yet subsequent experiments confirm the theoretical predictions. Figure 2 shows the first such example for a molecular target.

The electron-molecule calculations typically rely on the Born-Oppenheimer approximation that separates nuclear motion from the electronic motion. The treatment of vibrational levels can be done assuming an adiabatic treatment that combines the results performed at different internuclear separations. For diatomic molecules a spheroidal

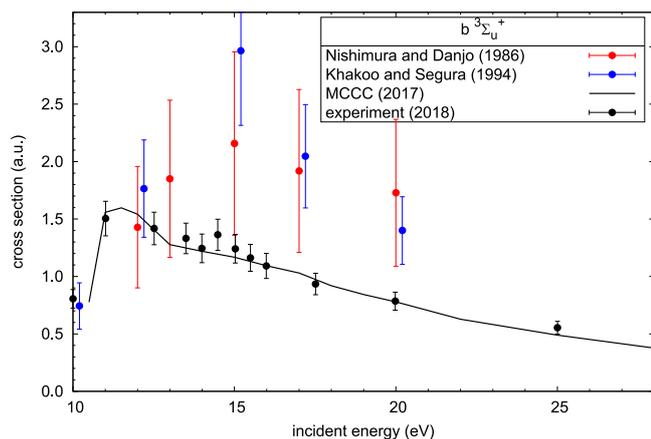


Figure 2. e-H<sub>2</sub>  $b^3\Sigma_u^+$  excitation see [8] for details.

coordinate system may be particularly advantageous to accurately treat the higher vibrational levels.

Not all aspects of electron–molecule scattering may be considered as solved, even for simple molecules such as H<sub>2</sub>. At sufficiently low energies coupling between vibrational and electronic excitation (even if only virtual) can be important.

*Future: experiment.* The great strength of the electron scattering field is the very close interaction between theory and experiment. We see no reason why this should be any different in the future. We acknowledge that the considerable progress in the field has resulted in fewer groups who are able to perform the kind of measurements that previously routinely challenged theory. The theoretical progress for the few-electron atoms and molecules would not have been possible without the experimental support. In the same way progress for more complex atomic and molecular targets will not be possible without the associated experimental support. As far as we are aware the last major discrepancy between experiment and theory for simple targets has been recently resolved [9]. As argued above, in the future the motivation will be more around studying those atomic and molecular targets that have practical applications. The interplay between theory and experiment will be much the same. Experiment would set a few benchmark measurements for specific cases. Once theory is able to reproduce the experiment the theory could then be applied more broadly to provide the extensive data required in applications. Arguably one of the most important molecules is H<sub>2</sub>O, whether in the gas or liquid

phase. Interactions with electrons have many applications ranging from atmospheric modeling through to medical therapy.

*Future: theory.* The goal of theory is to produce a broad set of accurate data for use in applications. Theory needs to be able to model targets sufficiently accurately in a way that subsequent scattering calculations can be made. Successful implementation for electron scattering means that the same target computational development can be used for other projectiles such as positrons or (anti)protons. The CCC method has evolved utilizing this principle [10].

With the aforementioned theoretical progress, the focus now and into the future is on multi-electron atoms and molecules. This means that considerable extra computational resources will be required compared to what has been available in the past.

*Computer code enhancement using graphics processing units (GPUs).* There is a quiet revolution going on in the field of supercomputation. GPUs are much more energy efficient and faster for floating point arithmetic than the more traditional CPU cores. Their historic slowness in data transfer has been addressed, and new computers are becoming available with many CPU cores and GPUs. This creates an opportunity for three levels of parallelism: between computer nodes, cores and GPUs. General computer codes intended to calculate electron interactions with complex atoms and molecules would benefit substantially from this hardware and associated software development.

*Concluding remarks.* Electron scattering theory has seen immense progress in the last few decades, particularly for quasi one- and two-electron targets. In part this is due to growth in computational resources, but also due to formal theory development. Atomic and molecular targets of interest to science and industry will dominate future applications, with recently developed supercomputer hardware being a critical enabler of further theoretical progress. The interplay of theory and experiment needs to be as strong as ever.

## Acknowledgments

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## 2. Polarization, alignment, and orientation in atomic collisions

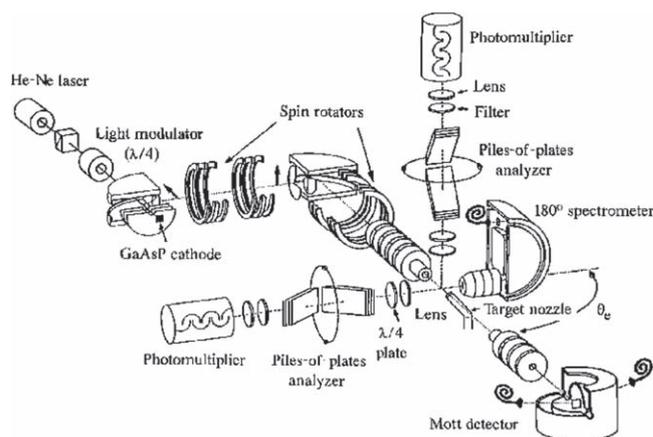
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*Status.* While cross sections are the key ingredients for many modeling applications in atomic collision physics, it is the study of polarization, alignment, and orientation (PAO) effects that probes the collision dynamics in greatest detail. To begin with, the linear polarization of the light emitted after impact excitation by a directional beam of charged particles (often, but not always electrons) without observation of the scattered projectile, corresponding to an angle-integrated Stokes-parameter measurement, revealed the ‘alignment’ of the excited state. Since those early days of Skinner and Appleyard [11], the field has advanced tremendously, often allowing angle-differential studies. Benchmark experiments were performed, first with unpolarized incident-particle beams, then with spin-polarized initial beams with or without analysis of the spin polarization after the collision, electron–photon coincidence experiments for inelastic (excitation) processes as well as (e, 2e) ionization setups, some of which resulted in excited target states that could optically decay and, once again, provide additional information through the polarization of the emitted light. Figure 3 shows an apparatus used in Münster in the early 1990s [12], where many such experiments were performed in the group of J Kessler and G F Hanne. In several cases, the so-called ‘complete experiment’ was realized, in which the complex-valued scattering amplitudes (magnitudes and relative phases) were determined. This is the maximum information that can be extracted according to Quantum Mechanics, and hence such benchmark experiments served as the most detailed tests of rapidly-advancing theoretical descriptions.

In addition to electron impact, heavy-particle as well as photon-impact studies developed in this area. The state of the valence electrons of the heavy particles could be prepared before and/or analyzed after the collision, thereby providing a rich playground for studying the collision dynamics. Advances in light sources, moving from relatively simple discharge lamps to synchrotrons, free-electron lasers (FELs), and few-cycle intense infrared (IR) sources, have further opened up the field. At the same time, advances in detector design (e.g. the reaction microscope [13] and the magnetic angle changer [14]) helped tremendously. An overview of the current status of the field can be found in the recent book by Andersen and Bartschat [15]. In addition to the latest developments at the time of publication, references to other monographs, extensive reviews, and pioneering papers that shaped the field over nearly a century, can also be found there.

As a result of the close collaboration between experimentalists and theorists, it is fair to say that relatively simple atomic targets (H, He, light alkalis and alkaline-earth elements) are now well understood. Theorists, of course,



**Figure 3.** Experimental setup of Sohn and Hanne [12]. © IOP Publishing Ltd. All rights reserved.

took advantage of the rapidly-evolving computational techniques and facilities, which allowed new (as well as sometimes old) ideas to finally be implemented into large suites of computer programs. Due to the limited space here, we suggest [15] for further references.

*Current and future challenges.* Given the excellent foundation established by the now very satisfactory agreement between experimental benchmark data and predictions from highly sophisticated numerical calculations for simple targets, new challenges can be addressed. In spite of the progress made for high-*Z* atomic targets such as Hg, heavy systems with open shells in the initial and/or final state remain problematic. It is not clear whether using a semi-relativistic approach based on the Breit–Pauli Hamiltonian (essentially a first-order perturbative correction to account for relativistic effects) is sufficient, or whether a full-relativistic treatment based on the many-electron Dirac equation is necessary. The accuracy of theoretical predictions for electron collisions with Xe or Pb, to name just two examples, remains questionable. The structure description is already highly challenging, and hence it is by no means straightforward to assess the quality of the collision model. Furthermore, the number of coupled channels increases drastically in a relativistic coupling scheme compared to a nonrelativistic treatment (and again by a factor of two when Dirac rather than Breit–Pauli is used), and hence the convergence of even the best currently available close-coupling models is difficult to establish. Finally, some open questions remain. One example concerns spin-polarized electron impact on Zn, where experimental results from different groups vary dramatically, and hence at best one set can agree with theory. See [16] for details and further references.

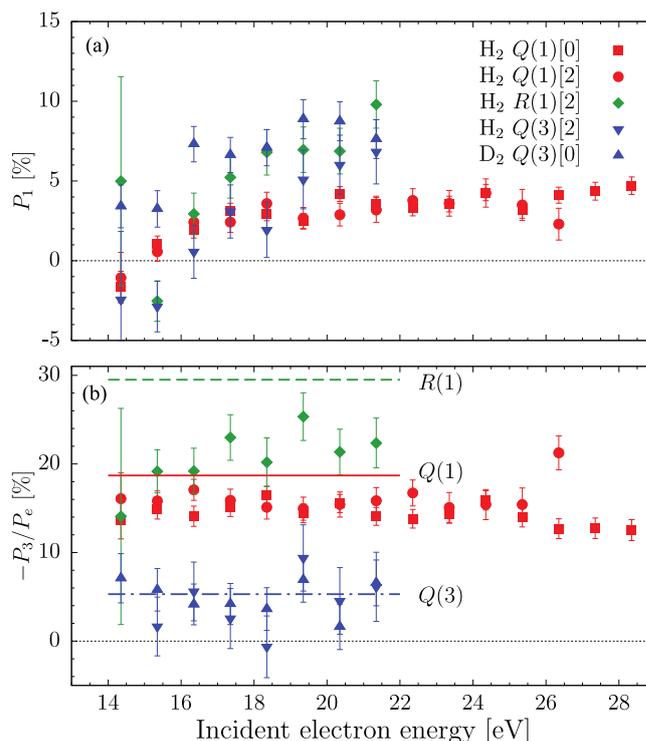
Another major challenge, for both experiment and theory, occurs when the single-center atomic targets are replaced by multi-center molecules. Experimentally, rotations and vibrations complicate the interpretation of the results, which may depend on the orientation of the molecule as well as the nuclear distance when away from equilibrium. At the same time, the numerical methods used for molecular targets

are (not surprisingly) lagging behind those for atoms. Even if the Born–Oppenheimer approximation is valid, the multi-center nature of the molecules either slows down the convergence of single-center expansions dramatically or causes additional complications when this simplicity is given up. For light diatomic molecules, with  $H_2$  being the simplest target for which PAO studies have been performed, some light is on the horizon. In general, however, there is a lot of room for further improvement.

Figure 4 shows one example of a PAO study on a molecule. Spin-polarized electrons excite various transitions in  $H_2$ , and the polarization of the emitted radiation is observed—here without detecting the scattered electrons [17]. While an approximate analytic model for the circular polarization is possible, predicting the linear polarization requires a dynamical treatment. Such a calculation is currently not available.

While being a topic originally developed in atomic collision physics, and here described mostly in the context of electron collisions, it is worth mentioning that polarization, alignment, and orientation are by no means limited to this field. Details can be found in Chapter 12 of [15]. Among other topics, we mention here Auger decays, collisions with surfaces, plasma polarization spectroscopy, spin-polarized beams for nuclear and particle physics, and—last but not least—quantum entanglement and Bell correlations. While it remains an open question whether or not the suggestion of creating a tunable entanglement via electron exchange collisions [18] will ultimately even lead to technological applications, it is certainly an interesting new twist to the field.

*Advances in science and technology to meet challenges.* As mentioned already, major advances in sources (e.g. FELs that can deliver strong polarized radiation over a large frequency range) and detector technology (e.g. the reaction microscope and the magnetic angle changer) have occurred. We expect further progress in these areas, which will benefit the efficiency and accuracy of the complex experimental studies. Regarding the theoretical side, algorithm development, coupled with the continuing rapid advances in computer technology, is expected to enable even more sophisticated calculations to be carried out than what is currently possible. In addition to solving the time-independent Schrödinger (or Dirac) equation more accurately, time-dependent approaches are being constantly improved. They are often needed to handle both heavy-particle collisions and short-pulse, intense laser-matter interactions. As one recent example, we mention the study of FEL-driven ionization, followed by excitation of the residual  $He^+$  ion by the same pulse, followed by multiphoton ionization of the excited ion—all by simultaneously using



**Figure 4.** Linear (a) and circular (b) polarization fractions for the  $H_2$   $d^3\Pi_u \rightarrow a^3\Sigma_g^+$  emission band. The circular-polarization data were normalized to the incident electron polarization and can be predicted approximately by using purely angular-momentum coupling [17], as shown by the straight lines. Reprinted with permission from [17], copyright 2013 by the American Physical Society.

circularly-polarized FEL and IR radiation with equal or opposite helicities to study circular dichroism [19].

*Concluding remarks.* The study of polarization, alignment, and orientation remains the key to understanding the dynamics of atomic collisions. For electron scattering, light quasi-one and quasi-two electron targets are now well understood. Hence, the challenge has moved to complex heavy atomic targets and molecular systems. Much progress can still be expected in studies involving heavy particles and particularly next-generation light sources. Also, the field continues to be relevant in applications that require more than cross sections for analyzing or modeling, and it may reach out into new areas such as quantum information.

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### 3. Electron-impact ionization of laser-aligned atoms

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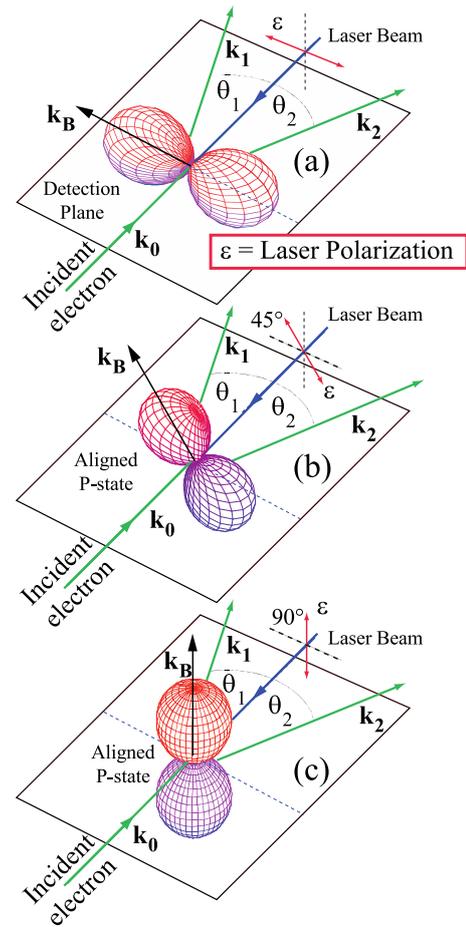
**Status.** Understanding electron-impact ionization of atoms and molecules is important in areas from astrophysics to Tokomaks, as detailed in the ‘Roadmap’ articles published here. Almost all experiments have been conducted on ground-state targets, however it is also important to understand these interactions from *excited* targets, since under certain conditions (e.g. in stellar atmospheres and in cooler plasmas), a significant fraction of interacting targets may be excited thermally or through collisions. Until recently measurements from excited targets has been challenging, since their production is difficult. As such, there is an almost complete absence of experimental testing of models that predict excited-state collision processes. Since these calculations are used in plasma modeling (e.g. in heat-loss predictions in Tokomaks), it is important to test them to ascertain their accuracy, and allow them to be refined if required.

The first (e, 2e) measurements from excited atoms were conducted by the Flinders group [20], who used circularly polarized laser light to create an ensemble of sodium atoms oriented in the scattering plane. The atoms were then ionized by electron collisions to study a prediction of chirality in the scattering process, the data agreeing well with the model.

Studies from *aligned* alkali atoms are considerably more difficult, since their hyperfine structure reduces the efficiency of the alignment process. By contrast, most alkaline-earth targets can be aligned with nearly 100% efficiency since they have zero nuclear spin. Their laser-excited P-states can then be aligned in different directions with respect to the collision frame (see figure 5), thereby producing a four-fold cross section (QDCS) that depends on the incident electron momentum  $\mathbf{k}_0$ , the scattered and ejected electron momenta ( $\mathbf{k}_1$ ,  $\mathbf{k}_2$ ), and the alignment of the excited atom  $\mathbf{k}_B$ . Comparison of the measured QDCS to theoretical calculations can then be made [21–24].

**Current and future challenges.** The first measurements from aligned atoms were conducted from  $^{24}\text{Mg}$  and  $^{40}\text{Ca}$  targets, which required radiation at  $\sim 285.3$  nm and  $\sim 422.7$  nm respectively. The light was produced from frequency-doubled continuous-wave (CW) lasers that could be tuned to an accuracy of better than 1 part in  $10^9$ . The lasers had to deliver at least 50 mW of power, and had to be stable for the length of time the (e, 2e) coincidence experiments operated (up to several weeks).

It is only recently that the laser systems required for these experiments have become available. For Mg studies, a Spectra Physics Matisse CW dye laser produced  $\sim 1.5$  W of radiation at 570.6 nm, which was then frequency-doubled in a Wavetrain enhancement cavity. The radiation was transported to the experiment via UV optics before being polarized by a BBO Glan-laser polarizer. The polarization vector (and hence  $\mathbf{k}_B$ ) was then rotated using a zero-order  $\lambda/2$  plate. Radiation for Ca excitation was provided by a Matisse Titanium



**Figure 5.** Alignment of the excited P-state with respect to the detection plane spanned by  $\mathbf{k}_1$  and  $\mathbf{k}_2$ . Three examples are shown, with the state aligned in the plane, at  $45^\circ$  to the plane and orthogonal to the plane.

Sapphire laser producing  $\sim 2.5$  W at 845.4 nm, which was again frequency-doubled in a Wavetrain cavity. As for Mg, the light was transported to the apparatus, polarized by a Glan-laser polarizer and the polarization vector adjusted using a zero-order  $\lambda/2$  plate.

Two types of experiment were conducted from excited  $^{24}\text{Mg}$  in these studies. In the first the laser was injected in the plane (as in figure 5), and in the second the laser was injected orthogonal to the plane so that  $\mathbf{k}_B$  rotated in the plane. In both cases a coplanar geometry was chosen, where  $\mathbf{k}_0$  was in the detection plane spanned by  $\mathbf{k}_1$  and  $\mathbf{k}_2$ , and  $\mathbf{k}_1$  was fixed at  $30^\circ$ . For experiments from excited  $^{40}\text{Ca}$  targets, only the geometry in figure 5 has been adopted up to the present, with  $\mathbf{k}_1$  fixed at  $45^\circ$ .

The QDCS calculated when  $\mathbf{k}_B$  was rotated in the plane [22] was compared to a three-body distorted wave model (3DW) and distorted wave Born approximation (DWBA) in [23], and reasonable agreement was found between theory and experiment. A parameterization of the QDCS formulated in [22] was then shown to be exact by Stauffer [25].

By contrast, when the excited atoms were aligned out of the plane, a large disagreement was found between experiment and the models. This disagreement was particularly

large when  $\mathbf{k}_B$  was orthogonal to the plane (as in figure 5(c)), since both 3DW and DWBA models predict the QDCS should be identically zero under these conditions, in marked contrast to the experiment [21]. Further work by the theoretical group of Colgan and co-workers at Los Alamos labs showed that inclusion of *un-natural parity* states to the calculated cross section will result in a finite measurement in this geometry [24].

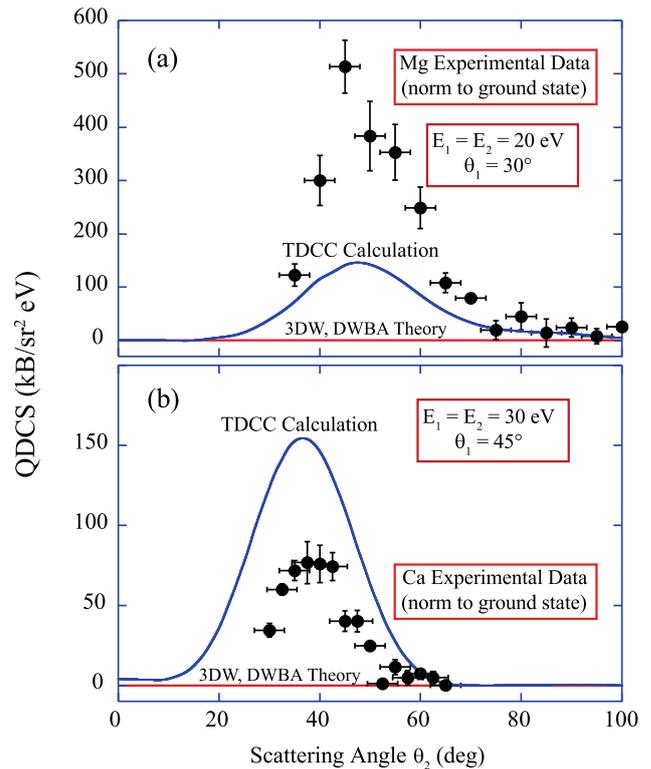
Figure 6 compares the data to the models for both Mg and Ca when  $\mathbf{k}_B$  is orthogonal to the scattering plane. By including un-natural parity states in the calculation, the TDCC model predicts a finite cross section whose peak position agrees well with the data. For Mg the magnitude of the QDCS is underestimated, whereas for Ca it is overestimated. Both 3DW and DWBA models disagree with the data, since they do not include these un-natural parity states.

#### Advances in science and technology to meet the challenges.

It is clear from the measurements shown in figure 6 that further work is needed to resolve the discrepancies that are found between the different models and experiment. The measured QDCS under these conditions are substantial, and are found to be  $\sim 50\%$  of the ionization cross section from atoms in the ground state. It is hence important for the models to be improved, so that correct cross sections can be calculated.

Further experiments are now underway to provide additional data for these ionization processes from excited targets. The experiments carried out so far have only considered a coplanar geometry where the outgoing electrons carry equal energy, and where one of them is fixed in space. Many other kinematic conditions are possible, including unequal energy configurations, as well as measurements where the incident electron is taken out of the detection plane. Such non-coplanar geometries have provided new insights into ionization from ground state targets [26–28], and are expected to also reveal new information from excited atoms. Modifications of the spectrometers in Manchester are currently underway to allow these types of measurement to be conducted.

From an experimental viewpoint, one of the key challenges to this type of experiment is to stabilize the laser system over the long time period required for coincidence measurements. To resolve this challenge a new type of control system has been developed that links the laser directly to the software that controls the (e, 2e) spectrometer. Tunable CW lasers are extremely sensitive to vibrations and temperature changes that cause them to mode-hop, and these new systems allow the laser to be retuned and optimized automatically by the spectrometer software, and then relocked to the correct transition required for excitation of the target. This is a step-change in laser control that then makes possible the more challenging experiments envisaged here. These controls will allow new experiments to be conducted that provide the data needed to resolve the current discrepancies between theory and experiment as discussed above.



**Figure 6.** QDCS when  $\mathbf{k}_B$  is aligned orthogonal to the plane for (a) Mg and (b) Ca. Both 3DW and DWBA theories predict QDCS = 0 for all scattering angles. The TDCC calculation which includes un-natural parity states predicts a non-zero cross section, although the magnitudes do not agree with experiment. The data are normalized to results for ground state ionization.

**Concluding remarks.** Ionization studies from excited targets have finally become feasible due to the development of new laser systems and control technologies that allow these difficult coincidence experiments to be conducted. The first detailed measurements carried out from aligned atoms show significant discrepancies exist between current models and experimental data. Since the density of excited targets can be significant in different plasmas, these discrepancies need to be resolved for theory to confidently calculate the cross sections required for injection into plasma kinetics models. This is a new area of research that hence has importance from both a fundamental point of view and for understanding energy losses in future fusion reactors. The technologies now exist to carry out these types of experiments, and it will be enlightening to see what new information will be obtained from their study.

#### Acknowledgments

We would like to thank the EPSRC, UK for providing funding for this work through grant R120272. The Photon Science Institute is also thanked for providing the CW laser systems.

## 4. Electron collisions with molecules

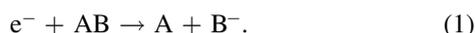
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**Status.** Electron collisions with molecules not only provide a means of probing structure and behaviour of molecules but are widely occurring. In nature they are found in lightning strikes as well as planetary aurorae and ionospheres; the top of the Earth's atmosphere and the tails of comets are bombarded by electrons from the solar wind. Cold plasmas, a state which involves partial ionisation of molecules, occur naturally in places ranging from flames to the interstellar medium. Astrophysically-important electron-collision processes are discussed in section 16 below.

Human technologies increasingly harness electron–molecules collisions: in the spark plugs that start cars, in traditional light bulbs, to activate many lasers and, probably most importantly, in the numerous technologies that etch and coat materials using plasmas. Plasma processing forms one of the major drivers of the modern economy. The resulting demands of scientists and engineers wishing to model molecular plasmas have led to the systematic compilations of electron collision datasets for key species (feedstock gases) such as  $\text{NF}_3$  [29], see figure 7 for example.

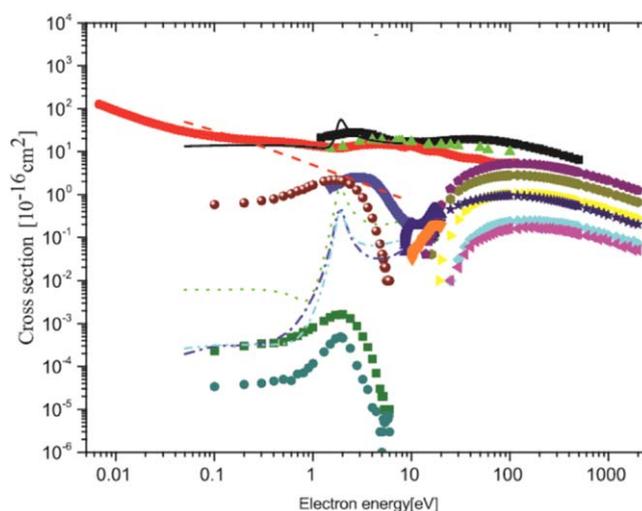
In the current century it has been realised that the damage experienced by bio-systems as a consequence bombardment by all types of high energy particles and radiation is predominantly caused by collisions involving low-energy, secondary electrons with individual biomolecules. Such collisions can be both harmful in that they cause double strand breaks in DNA [30] or potentially useful when employed as the basis for therapies such as cancer treatments. The key process in these collisions is thought to be dissociative electron attachment (DEA) [31]



The important DEA process is discussed in detail in section 7 below.

The measurement of electron–molecule collision cross sections is a mature field with, in general, a decline in this activity worldwide. Exceptions to this being the study of collisions with biomolecules [32], see section 9, for reasons discussed above and the study of novel phenomena resulting from collision processes [33], see section 6. The development of velocity map imaging, particularly for studies of DEA, has proved an important step in both these areas.

The levelling off or decline in experimental activity is not mirrored by theoretical work. Increased computer power has led calculations which can both be performed on larger systems and with increasing accuracy. As is usual, increased computer power has led to improved algorithms with matching benefits in terms of increased scope and accuracy. Two such algorithmic improvements are particularly noteworthy. The first is the use of quasi-complete expansions of target wave functions as represented by the CCC approach [33], see section 1 above. This method allows calculations



**Figure 7.** Summary of recommended cross section for electron collisions with the  $\text{NF}_3$  molecule: Thick black line—total scattering, green triangle—elastic scattering, thick red line—momentum transfer, thin lines rotational excitation (red  $\Delta J = 0$ , black  $\Delta J = 1$ , green  $\Delta J = 2$ , dark blue  $\Delta J = 3$ , light blue  $\Delta J = 4$ ), brown circles, green squares and green circles—dissociative electron attachment, thick blue line—vibrational excitation, other symbols represent various combinations of ionisation and dissociation. For further details see Song *et al* [29].

that bridge the challenging intermediate energy region which spans the gap between low energy collisions, which occur below the target molecules ionisation threshold, and high energy collisions above a 100 or so eV, where perturbation theory methods can be considered reliable. The effective complete treatment of the target electronic states also facilitates a comprehensive treatment of the scattering electron–target electron correlation problem, often called polarisation, resulting in greatly improved accuracy [33].

The second advance is the use of B-spline basis functions to provide a flexible and extensible representation of the continuum [34, 35]. Use of these functions allows the treatment of much more extended systems than have been studied heretofore: meaning, in principle, both large molecules and molecules with extended or diffuse excited states can also be studied. As the electronic excited of all molecules rapidly become diffuse or Rydberg-like, this property is important for studies of electronic excitation.

**Current and future challenges.** As discussed above, the study of electron–molecule collisions is relatively mature but there remains major drivers which demand further studies.

Both experimentally and theoretically, the studies of electron collisions with bio-systems with a particular emphasis on DEA has proved a major driver in the discipline [31, 32]. However, the overwhelming majority of this work has concentrated on the low-lying shape resonances that are displayed by various DNA fragments and related molecules; most of these studies have concentrated on resonance lying below 5 eV. Yet the initial pioneering experiments of Boudaiffa *et al* [30], and subsequent work by the same group, strongly suggests that the critical energy at which

much of the damage occurs is at slightly higher, in the 5–10 eV region. In this region most of the resonances are Feshbach in nature which makes any theoretical treatment significantly more demanding. Furthermore, biomolecules do not exist in isolation in living systems. The role of hydration on the various resonant processes needs to be more thoroughly explored.

While compilations of validated or recommended data are very useful for modellers, the current compilations focus almost exclusively on electron collisions with stable, closed-shell molecules. The species are amenable to experimental study and this, combined with appropriate theory and swarm studies, can lead to reliable and fairly comprehensive datasets, such the one illustrated in figure 7. However, plasmas are chemically active environments containing many radicals (open shell species) and molecular ions. Experimental studies of electron collision with radicals, in particular, are almost non-existent. This has led to the assumption that theoretical methods will provide the future workhorse when it comes to providing the complete datasets that plasma require [36].

This assumption is probably correct. However, if computation does represent the best hope of providing bulk data, there are particular processes where better validation of theoretical methods by experiment is important. The first is electron collisions with radicals: just because a procedure gives reliable predictions of cross sections for electron collisions with a set of close-shell molecules, there is no guarantee that use of the same procedure will give equally good results for electron collisions with a radical. Second, electron impact vibrational excitation is increasingly being recognised as an important driver in many cold plasmas and there is an urgent need for reliable vibrational excitation cross sections for species such as CO<sub>2</sub>, which cover an extended range of vibrational states and transitions. Experimental studies on electron-impact vibrational excitation for polyatomic molecules remain limited. Thirdly, electron impact rotational excitation of molecules, particularly ones with large dipole moments, is being increasingly recognised as an important process in partially ionised regions of the interstellar medium such photon-dominated regions (PDRs). Calculation of these cross sections are fairly straightforward but there is a lack of experimental validation of these calculated results.

#### *Advances in science and technology to meet challenges.*

The increasing use of electron–molecule collision cross sections in models has raised the issue of the reliability to which these cross sections can be obtained. While experimentalists have a long tradition of providing their

data with appropriate uncertainties, the same is not true for theoreticians. Protocols for uncertainty quantification (UQ) which can be applied to computed cross-sections have been proposed [37], but, as yet, have only been very rarely applied. Of course use of these protocols implies repeat calculations and increased computation times, but as theoretical models improve so will the need to supply realistic and usable uncertainties.

Pseudo-state methods, such as CCC discussed above or the more flexible but probably less formally rigorous *R*-matrix with pseudostate method (RMPS), actually offer the possibility giving UQ; indeed CCC calculations on the electron–H<sub>2</sub> problem represent one of the few cases where computed results have been presented with uncertainties [38]. The challenge is to extend such calculations, including UQ, to larger problems of importance.

The key species forming important constituents in technological plasmas are becoming increasingly complex. B-spline-based procedures should be capable of treating electron collisions with large molecules but thus far have only been applied to electron collisions with two [34] or few [35] electron targets. While B-splines work well for the limited applications for which they have been used, extending the methodology to the study of the general, multi-electron problems remains challenging because of the expense of computing all the relevant integrals

The compilation of complete datasets for electron collisions with key species provides an important service for the user community who frequently lack the skills to identify which, from a range of published cross sections, provide the best data for use in models. Most of these studies are published on a molecule-by-molecule basis. A start has been made in providing aggregated databases through projects such as the Virtual Atomic and Molecular Data Centre (VAMDC), LXCat and the Quantemol DataBase (QDB). For this activity to flourish these compilations will need to move from aggregating what is available towards ensuring that entire sets, sometimes called chemistries, are provided for the modeling community.

*Concluding remarks.* There are well established procedures for measuring electron–molecule collision cross sections which are being complemented or even supplanted by increasingly accurate and extensive theoretical methods. The direction of this work is being increasingly driven by the needs of user communities which includes studies of radiation damage in living systems, astrophysical processes or the desire to make accurate models plasma processing.

## 5. Kinematically complete ionization studies

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**Status.** A full understanding of the correlated fragmentation dynamics of an atomic system upon impact of a charged particle or a photon is one of the central aims of atomic collision physics. Very detailed insight give kinematically complete or (e, 2e) studies which determine the momentum vectors of all continuum particles in the final state. (e, 2e) experiments became feasible already very early in the late 1960s more than two decades before the advent of kinematically complete experiments for ion impact ionization (for a review of the early work see [39]). In the standard experimental technique both outgoing electrons—the scattered projectile and the ejected electron—are detected in coincidence with electrostatic analyzers positioned mostly in one plane including the incoming projectile beam (coplanar geometry). The angular distribution patterns of the observed electrons can be interpreted in terms of relevant interactions and reaction mechanisms and serve to test theoretical calculations. In course of time a large body of studies where performed covering diverse gaseous targets (but also solids) and various kinematical situations including relativistic collisions, spin polarized projectile beams and laser excited targets. Guided by experiments theory showed tremendous advances as the development of various versions of distorted wave approximations (DWA) and analytical three-body wave functions (3C) to name just two approaches. Perturbative models can give insight in the importance of specific two-body interactions by alternatively including or omitting them. This was nicely illustrated in a study by Al-Hagan *et al* who identified higher order projectile-target interaction to result in emission of both outgoing electrons perpendicular to the incoming projectile beam [40]. Eventually the increasing computer power enabled the development of non-perturbative methods like convergent-close-coupling (CCC), time-dependent close-coupling (TDCC) [41] and exterior complex scaling (ECS) [42] which have demonstrated to give essentially exact cross sections for the most fundamental targets like atomic hydrogen and helium. Larger atoms can be treated with the B-spline *R*-matrix method (BSR). Experimentally, in the 90s of the last century more efficient multi-parameter coincidence spectrometers [43] and multi-particle imaging techniques as the reaction microscope (ReMi) [13] were developed with strongly increased efficiency due to large solid angle coverage and the acceptance of a range of ejected electron energies. In addition to the electrons the ReMi records the recoil ion giving additional information, e.g. on the fragmentation of residual molecular ions.

**Current and future challenges.** While for the most basic three-body breakup processes such as pure single ionization a rather profound understanding has been achieved the transition to true many-body systems still remains a challenge, both experimentally and theoretically. With the availability of experimental multi-coincidence techniques

complete momentum space pictures of complex many-body processes can be obtained. One example is double ionization of helium by electron impact leading to four unbound particles showing intriguing and diverse dynamics in going from high impact energies to the threshold region. For these (e, 3e) reactions theory is still in its infancy and for a long time could not treat all mutual interactions on an equal footing. Only recently consistent non-perturbative results using the above mentioned TDCC model were published by Colgan and coworkers [44]. A particular open question concerns the emission pattern of the three electrons close to the ionization threshold. Recent classical and quantum mechanical calculations predict a symmetric triangular emission for ionization of equivalent electrons but surprisingly a T-shape pattern if electrons in different shells are ionized. This is in contradiction to the successful and generally accepted Wannier model which treats the emission to be independent of the target structure and completely determined by the particle correlations in the continuum.

Another fundamental four-body reaction is the simultaneous ionization and excitation of helium. Here an (e,  $\gamma$ 2e) experiment, i.e. a coincidence with the photon emitted by the excited ion and the analysis of its polarization is required to fully characterize the excited ionic state. This process is very interesting in particular at low impact energies. Similar to double ionization it involves two active target electrons but since there are only two continuum electrons in the final state their angular emission pattern is less dominated by long range post collision interaction allowing better insight in the collision dynamics at short distances. While triple coincidences studies were done with both existing ReMi setups with implemented electron projectile beam in Lanzhou and in Heidelberg a polarization or angular distribution analysis of the photons could not be realized up to now.

If ionization takes place in a strong laser field the collision dynamics can be strongly modified and for high intensities a perturbative description fails. This is the case, e.g. for the non-sequential double ionization in intense laser pulses where an initially ionized electron is re-colliding and further ionizing the parent ion. The observation of this reaction under controlled conditions in a dedicated experiment is difficult and there exists only one successful attempt for a so-called ( $n\gamma$ e, 2e) experiment on helium atoms [45].

Presently (e, 2e) studies for electron impact mainly deal with targets of increased complexity. These are molecules on one hand and clusters on the other hand. Molecular targets are challenging for theory due to their multi-center nature and spatial anisotropy which cannot be considered in an exact way by present day methods. Nevertheless, reasonable results are obtained by various models like the molecular 3-body distorted-wave approach (M3DW) and others. Naturally the best results are obtained for the simplest molecule  $H_2$  for which nowadays non-perturbative TDCC and CCC calculations are feasible. Regularly the molecular multi-center structure is described by a linear combination of atomic orbitals and electron ionization by a coherent superposition of single center waves emitted from the individual atoms. This treatment neglects the multi-center effects on the outgoing

waves. As result for two-center targets like  $H_2$  the well-known Young's double-slit interference is obtained for electron emission which is discussed in the fields of ion and electron impact since a seminal work by Stolterfoht and coworkers in 2001 [46]. Most existing studies which claimed the identification of this interference pattern averaged over the molecular spatial alignment. Current ongoing experiments can identify the alignment for collisions where the residual ion dissociates. First results demonstrate that at low ejected electron energies the collision dynamics is rather complex and Young's interference is not visible. Future alignment resolved studies will aim for higher energies of the emitted electron where the prerequisites for the observation of the double slit interference are expected to be fulfilled.

Topical target species include such of relevance in nature and for technical applications. One example is the ongoing and intense research on radiation effects in biological tissue. Here one of many more goals is to maximize damage to cancer cells in radio therapy treatment, e.g. with help of radio sensitizers. Therefore, ionization cross sections for the slow secondary electrons which are abundantly produced by the primary particle are required for example for track structure simulations. Here it is unclear if the presently solely used single and double differential cross sections are sufficient or if the usage of fully differential cross sections would cause significant differences in the obtained track structures, the ionization patterns and the clustering of damages. Another motivation is to better understand the underlying reaction mechanisms leading to radiation damages. Here, (e, 2e + ion) experiments where both outgoing electrons and the fragment ion are detected can contribute. These allow correlating the ionized molecular orbital with the subsequent dissociation in a similar way as it is done for photoionization with the successful PEPICO technique. In a respective (e, 2e) study on a model system for a hydrated biomolecule the intermolecular Coulombic decay mechanism (ICD) could be identified. In ICD the internal energy of an ionized water molecule is transferred across a hydrogen bridge to a neighboring tetrahydrofuran molecule ionizing it as well [47] and giving rise to a Coulomb explosion.

Such (e, 2e) studies on clusters which are produced in low abundance in a supersonic gas expansion and other thin targets become accessible due to the high efficiency of the ReMi technique. As result also the ionization dynamics in small aggregates of atoms can be addressed. In a measurement on argon dimers the electron impact ionization of both

atoms was observed by the coincident detection of all three electrons and both ions in the final state. Following the analysis of the kinetic energies two ionization mechanisms could be identified and separated. These are the above mentioned ICD and furthermore a radiative charge transfer process [48]. Earlier studies of the (e, 2e) angular distribution patterns on larger Ar clusters showed modifications with respect to the atomic target which can be understood in terms of multiple scattering processes including such which combine ionization of one atom and excitation of another atom.

*Advances in science and technology to meet challenges.* On the theoretical side there are ongoing efforts and advances. This is the case for perturbative approaches which now start, e. g., to consider the multi-center molecular potential without spherically averaging. For non-perturbative methods smaller molecules come into reach. Experimentally there is progress in various directions like the detector efficiency which can be increased from 54% to 83% with newly developed micro-channelplates [49]. For 5-particle coincidence this results in almost one order of magnitude higher rates. On the other hand the mediocre electron energy resolution of the ReMi technique at higher electron energies cannot be overcome generally but must be addressed in individual measurements for example with deceleration of the emitted electrons. In addition target sources providing sufficient densities for larger non-volatile and temperature-sensitive (bio-) molecules are required. Here techniques like laser induced acoustic desorption (LIAD) or matrix-assisted laser desorption (MALDI) are options to be tested.

*Concluding remarks.* The selected examples of early and current research discussed above demonstrate that kinematically complete studies of electron impact ionization have matured and go far beyond the early investigations of the fundamental three-particle problem. The field continues to provide insight in correlated dynamical processes and to develop new theoretical methods. In addition it reaches out in new areas like to the condensed phase and to biology.

## Acknowledgments

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## 6. Electron momentum spectroscopy: from static to time-resolved

Masakazu Yamazaki and Masahiko Takahashi

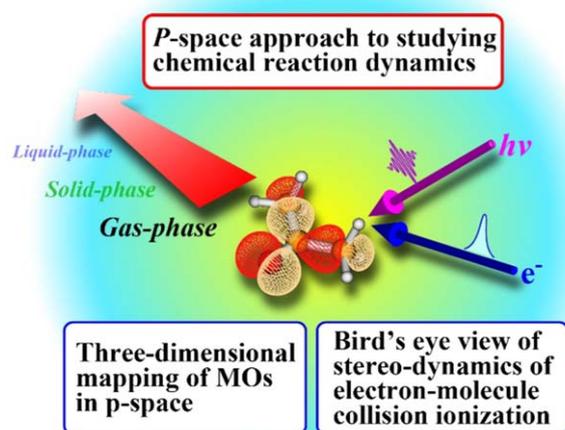
Tohoku University, Japan

**Status.** The ionization of atoms and molecules by electron impact is a test bed for the most fundamental laws in collision physics. It is also one of the most basic processes encountered in a broad range of research areas and applications, including atmospheric science, plasma science, radiation physics and chemistry, and biology. The extent to which such processes can be controlled and/or optimized is limited by our ability to describe the underlying physical mechanisms as well as the target electronic structure. There is thus an ever-increasing need for achieving a more complete understanding of electron-impact ionization. The most detailed information, triple differential cross section can be obtained with (e, 2e) spectroscopy, in which an incident electron having well-defined energy and momentum ionizes a target and the energies and momenta of the resultant two outgoing electrons are measured by a coincidence technique.

Electron momentum spectroscopy (EMS) [50, 51] is an experimental technique that is classified into (e, 2e) spectroscopy. The originality of EMS, however, lies in its capability to separately measure momentum distributions of each electron bound in a target or to look at individual electron orbitals in momentum space (*p*-space). Such measurements are possible if an (e, 2e) experiment is performed under the high-energy Bethe ridge conditions where the collision kinematics most nearly corresponds to a collision of two free electrons with the residual ion acting as a spectator. In other words, EMS can be recognized as an advanced form of traditional electron and x-ray Compton scattering experiments.

The history of EMS experiments is long and goes back to around 1970. The research streams developed for gas-phase atoms and molecules so far can be divided into the following seven topics;

- Electronic structure and electron correlation studies for a wide variety of targets ranging from atoms to molecules of biological interest [50, 51].
- Collision dynamics in electron Compton scattering, which has been investigated through comparisons between EMS experiments on two-electron processes and second-Born approximation calculations [51].
- Molecular frame EMS experiments [52], which have opened up two areas, i.e. three-dimensional mapping of molecular orbitals (MOs) in *p*-space and stereo-dynamics of electron–molecule collision ionization.
- EMS studies on distortion of MOs due to molecular vibration [53], which aim partially to elucidate the origin of vibronic coupling that involves interaction between electron and nuclear motions.
- Density oscillation, which is a phenomenon particular to *p*-space MOs and provides information about molecular



**Figure 8.** A roadmap for time-resolved electron momentum spectroscopy and related techniques.

geometry and spatial orientations and phases of the constituting atomic orbitals [54].

- High energy-resolution EMS experiments, involving observation of vibrationally-resolved electron momentum densities for H<sub>2</sub> [55].
- EMS experiments on laser-excited targets, which have been performed only for the three targets, the Na atom [56] and the acetone [57] and toluene molecules.

**Current and future challenges.** Great progress has been made in this field over the past 50 years and today there are more sophisticated researches underway than ever before. One sign of such constant challenges is development of time-resolved EMS (TR-EMS) [51, 57], which is an experimental technique that employs short-pulsed laser and electron pulses in a pump-probe scheme. It has very recently made it possible to conduct EMS measurements for short-lived molecules in their excited states with lifetimes of several to tens of picoseconds [57]. However, the technique of TR-EMS should be improved more and more for having much broader scope of future applications. Space does not allow the authors to talk about those all, so let them do about only three selections given below as well as in figure 8.

### (1) Three-dimensional mapping of MOs in *p*-space

EMS experiments usually suffer from the spherical averaging due to random spatial orientation of gaseous molecular targets. The pioneering work of three-dimensional mapping of MOs in *p*-space [52] and the following studies have solved the problem by additionally detecting the fragment ion produced through an axial-recoil fragmentation process of the residual ion. However, use of such processes limits the possible target molecules only to simple ones such as diatomic molecules. One of the strategies to overcome this

limitation is to combine TR-EMS with the technique of laser-induced alignment and orientation of gaseous molecules. Mapping MOs of various molecules freely in the three-dimensional form would have a far-reaching impact on many of molecular physics and chemistry. This is because various properties of a molecule such as chemical reactivity and molecular recognition are governed by its specific MO patterns, in particular their large  $r$  (small  $p$ ) parts, far from nuclei, to which EMS is highly sensitive.

(2) Bird's eye view of stereo-dynamics of electron-molecule collision ionization

The pioneering work of stereo-dynamics of electron-molecule collision ionization [52] and the following studies all have been performed for diatomic molecules and either at small or at large momentum transfers ( $K < 2$  a.u. or  $K \sim 10$  a.u.). This situation may recall the parable of the blind men and an elephant. It implies that subjective experience at small (large)  $K$  is true but may be inherently limited by its failure to account for alternate possibilities at different  $K$ 's or the sum total of facts. It is thus desired to take a bird's eye view of stereo-dynamics of electron-molecule collision ionization by covering a wide range of  $K$ . Furthermore, the use of molecules aligned or oriented by laser fields is a straightforward and excellent way to broaden the range of target species available. To these ends, the technique of TR-EMS will have to be tailored so that time-resolved versions of (e, 2e) experiments at various  $K$ 's can be conducted simultaneously. Since the change in  $K$  corresponds to the change in impact parameter, such a bird's eye view will show a full picture of what happens when a free electron having certain kinetic energy and momentum collides with and ionizes a target molecule, depending on, for instance, whether the collision is head-on or broadside and whether the collision is distant or close.

(3)  $P$ -space approach to studying chemical reaction dynamics

One of the goals in the field of chemical reaction dynamics may be to watch reactions in real time. Indeed, for instance, the time-resolved electron and x-ray diffraction technique has made it possible to visualize structural dynamics during chemical reaction. Nevertheless, there still remains the challenge, that is, to explore why the atoms are dancing in such a way. For this end, it is essential to develop a real-time spectroscopic complex that meets one's needs. One of such spectroscopic complexes desired is a set of TR-EMS and time-resolved version of atomic momentum spectroscopy (AMS) that is based on electron-atom Compton scattering [58, 59]. A more advanced use of the current TR-EMS technique [57] will make it possible to measure in real time the momentum

distributions of each electron, bound in a transient, evolving system, with different ionization energies, thereby enabling one to make a 'MO movie' in  $p$ -space. The observed change in electron motion would represent the driving force behind chemical reaction. On the other hand, the technique of time-resolved AMS (TR-AMS) is not present at this time, so it must be newly developed. It will be unique with its capability to measure the momentum distributions of each atom, involved in an evolving system, with different mass numbers, which tell about how and how much the change in atomic motions are brought about by the change in electron motion. A targeted goal of this real-time spectroscopic complex is to visualize the driving principle of a gas-phase photochemical reaction by providing a series of snapshot microscopic views about how and why the reaction proceeds in such a way.

*Advances in science and technology to meet challenges.*

What can be the most groundbreaking advance in science and technology, required for the challenges proposed here, would be an idea to solve the pump-probe velocity mismatch problem while keeping the incident electron beam current high enough to have a workable EMS signal count rate. A more intense, monochromated electron gun, a more intense molecular beam source, and a high transmission electron energy analyzer with much better energy resolution are always strongly required and desired. Finally, it is pointed out that application of TR-EMS to solid phase, beyond the early pioneering EMS studies on solids [50], and eventually to liquid phase will be feasible, as depicted in figure 8, when more advanced technologies to produce self-standing thin-film materials and a sufficiently intense cluster or droplet beam with thickness and average diameter of less than several tens of nm become available.

*Concluding remarks.* The research areas outlined here provide a glimpse of possible future efforts for EMS, which is currently extending its investigations from static to time-resolved studies. Upcoming trends are forecasted and the need to develop and improve experimental techniques is highlighted. The authors sincerely wish the present short paper could assist as many people as possible to find an interest in EMS.

### Acknowledgments

The authors are grateful to all the colleagues who have been and are involved in the field of EMS. They are also grateful to the financial support by the JSPS and MEXT as well as by the Cooperative Research Program of 'NJRC Mater. & Dev.'

## 7. Dissociative electron attachment—a route to chemical control

Nigel Mason

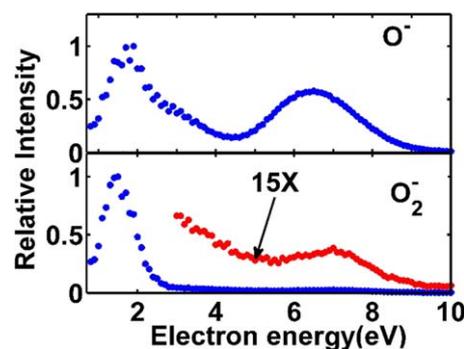
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*Status.* The process of dissociative electron attachment (DEA) is important in a range of scientific fields and technologies operating from large scale astrophysics to nanotechnology. DEA studies have therefore expanded in recent decades adopting novel experimental methodologies and exploiting computational advances to develop analogous theoretical studies [60].

DEA may be summarized as follows a low energy electron interacts with a molecule (AB) to form a short lived temporary anion  $AB^-$  which subsequently decays to form a fragment anion  $A^-$  and neutral products. The process is ‘resonant’ in that it occurs over a limited energy range (typically 100 meV to few eV) figure 9 and crucially is often bond specific. Thus each ‘resonance’ and temporary negative ion (TNI) may decay through a distinctive pathway rupturing one bond. Thus by altering the energy of the incident electron it is possible to select the molecular fragmentation pathway. Such ‘tunable’ fragmentation leading to specific fragments in turn can influence or even control local chemistry leading to the development of electron induced chemistry (EIC). EIC is now being invoked in plasma applications by altering the electron temperature in the plasma to tune concentration of radical species used in plasma etching. In next generation nanolithography DEA leading to the fragmentation of the molecular precursors is central to the development of focused electron beam induced deposition (FEBID) and the choice of photoresist substrates used in EUV lithography [61].

Perhaps the most dramatic example of the DEA in nature is the discovery that when low energy electrons interaction with DNA the subsequent patterns of both single and double strand breaks show distinct resonance features characteristic of the DEA patterns in the constituent molecules (e.g. nucleobases) [62]. This suggests, that in contrast to previous assumptions, it is not the primary ionizing radiation that causes DNA damage but the avalanche of lower energy secondary electrons. This in turn has led to a choice of radiosensitizers that show large DEA cross sections and the suggestion that metallic nanoparticles may be incorporated into cancer cells such that, when irradiated, they liberate low energy electrons which destroy DNA by DEA led processes killing the cell.

The recent Cassini–Huygens mission to Saturn revealed a preponderance of anions in the upper atmosphere of Titan due to DEA of constituent tholins ( $-CN$  containing compounds, known to be quasi halogens with large DEA cross sections [63]). Such anion processes may also be important in astrochemistry and DEA driven EIC may explain the rich organic chemistry found to occur on Comet 67P/Churyumov–Gerasimenko during the recent Rosetta missions [64].



**Figure 9.** Typical DEA anion yield curves demonstrated for  $O^-$  and  $O_2^-$  from ozone.

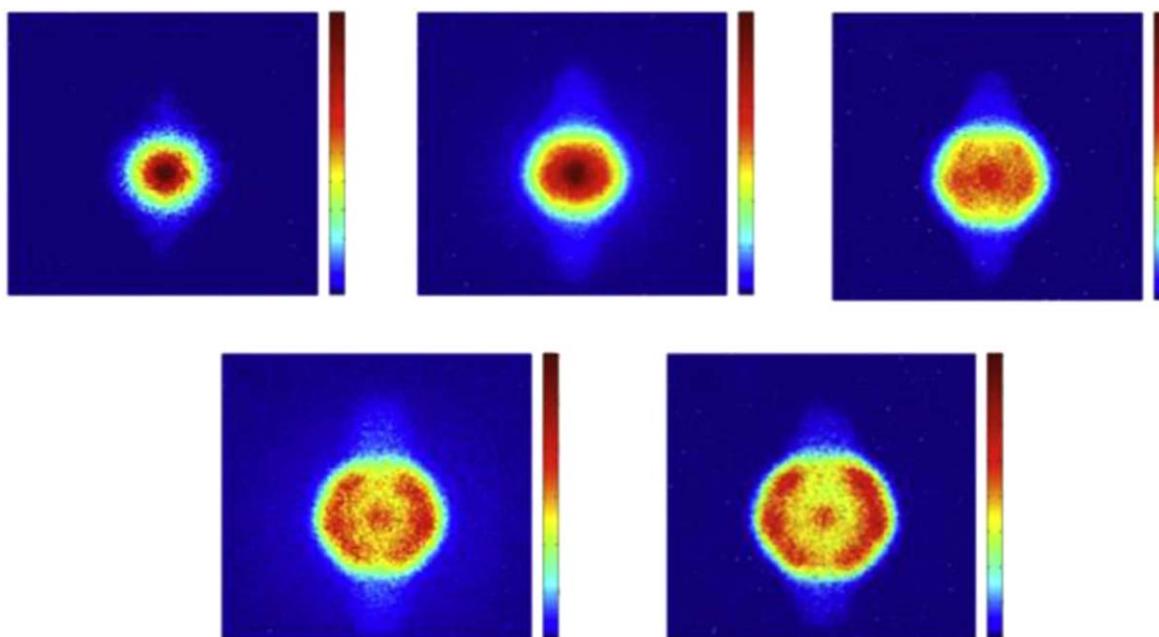
The detection of anions in the interstellar medium (ISM) also suggests new, largely unexplored, anion chemistry leading to creation of prebiotic molecules in the ISM themselves the potential origins of biochemistry leading to life on Earth (and other (exo)planets).

Thus DEA is a ubiquitous process whose study provides insights into many natural and technological phenomena. DEA is therefore currently one of the most active fields of atomic and molecular physics.

*Current and future challenges.* Understanding the dynamics of DEA and predicting/controlling its fragmentation pathways is a major challenge since at present we are unable to accurately predict the fragmentation patterns of DEA in any complex molecule. Indeed recent results have shown that our understanding of the DEA dynamics of even the simplest diatomic ( $H_2$  and its isotopes) is poor with unexpected quantum interference phenomena coupling decay paths and thence influencing the final fragmentation patterns [65]. Theoretical studies and simulations of DEA often fail to predict the experimental observations and DEA provides a major challenge to the electron–molecule scattering community.

DEA is prevalent in all phases of matter and is responsible for the chemical synthesis of molecules in ices and thin films under electron irradiation. However DEA processes, both cross sections and fragmentation pathways are strongly influenced by the local environment since the TNI is linked to neutral states of the parent molecule the excitation energy of which is shifted in dense and solid media. Indeed some TNI are suppressed completely. The current challenge is to determine methods for studying DEA dynamics with the same detail possible within the gas phase in condensed phase and determine the DEA cross section in such environments [66].

One method to explore the alteration of DEA as a function of phase is to explore DEA in molecular clusters. DEA may induced intra and inter-cluster chemistry revealing possible routes of molecular synthesis in condensed phase. However most cluster sources are an ensemble of different clusters dimers, trimers etc and the challenge is how to isolate



**Figure 10.** Velocity Map images of  $O^-/OH^-$  in acetaldehyde  $CH_3CHO$  recorded at 9.0, 9.5, 10.0, 10.5 and 11.0 eV showing two overlapping resonances distinguished by change in angular distributions [68]. Reproduced with permission of The Royal Society of Chemistry.

different clusters in an experiment to probe how DEA changes as a function of cluster size.

DEA must also occur in liquid phase but to date there are few electron studies with or in liquids (due to experimental challenges) and no comprehensive evidence for the observation of DEA induced phenomena in liquids. Developing an experimental methodology to explore (electron) scattering from liquids is therefore a core challenge to the molecular physics community.

It is also necessary to explore a related but currently understudied process that also produces anions, electron induced dipolar dissociation of Ion-Pair formation  $e + AB \rightarrow A^- + B^+$ . This anion production channel is particularly relevant in molecules for which DEA cross sections are low (e.g. hydrocarbons) since it is generic to all molecules and being non resonant will produce anions at all impact energies above a threshold energy.

#### *Advances in science and technology to meet challenges.*

The last decade has seen several major advances in experimental methodology used to study DEA. The adoption of Velocity Map (Slice) Imaging VMI/VSI, a method developed for study of photodissociation using lasers and synchrotron radiation, has revolutionized our knowledge of the dissociation dynamics of DEA [67]. Using VMI both the kinetic energy and the angular distributions of the product anions are measured using a combination of time of flight spectroscopy and spatial detection using a position sensitive detector (PSD). All of the anions with the same

velocity produced in a given interaction volume are mapped onto the same point onto the PSD. The image position on the PSD and the time to arrival define all three velocity components of the anion fragments when they are formed. A Newton sphere is created when each dissociation event yields two partner fragments travelling with equal momentum in opposite directions in the center of mass frame. When the same dissociation is observed a number of times, the fragments build up a spherical distribution in velocity space. The size of the image enables the kinetic energy of the ions to be derived, as the distance of the ion impact point on the detector from the centre of the image is directly proportional to the square root of the kinetic energy of the ion. Direct observation of the VMI image as a function of incident energy allows overlapping resonances and their symmetry to be easily distinguished (figure 10) [68].

Another significant advance has been the development of techniques to prepare new molecular targets in the gas phase including biomolecular targets for example using laser desorption systems or different forms of DNA, including the use of origami nanostructures [69], allowing electron scattering to be extended to macromolecules.

Considerable advances are being made in complementary theoretical calculations of DEA both identifying the position and lifetime of scattering resonances in larger scale macromolecules. Studies of how such resonances, and hence DEA cross sections, are altered by the local medium e.g. within clusters or condensed phase are

developing rapidly [60] and are essential if the role of such phenomena are to be assessed in biological and astronomical conditions.

*Concluding remarks.* DEA is an active area of current research with significant progress having been made in the last decade. The development of new experimental methods (VMI) coupled with advances in theoretical calculations allow DEA to ever more complex targets in all phases of matter to be explored. Such studies are revealing the intrinsic nature of

DEA across many fields of science and technology from astronomy to radiation therapy.

### **Acknowledgments**

NJM recognizes the support of several major collaborative research programmes exploring DEA across several research fields. EU funded programme LASSIE; VAMDC/Sup-VAMDC; ARGENT and ELENA and COST actions RADAM, ECCL, CELINA and the Chemical Cosmos.

## 8. Electron-ion recombination with atoms and cold molecules

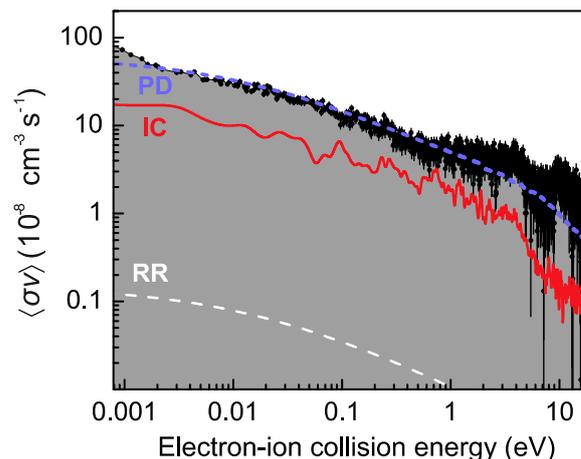
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**Status.** Plasmas and weakly ionized gases occur with a wide variety of densities, temperatures and chemical composition in space, planetary atmospheres and technical devices. These media hold potential energy through the separation of charges, which is dissipated when positive ions recombine with free electrons. The media are often dilute enough that binary collisions [70] dominate so that the recombination is described by cross sections as functions of the collision energy and by first-order rate constants as functions of the plasma temperature. These are key quantities for predicting the media properties, such as the abundance of high-charge state ions and the energy content in fusion plasmas or the effects of ionization on the chemical composition of cold interstellar molecular clouds, planetary discs, and planet atmospheres [71].

Theoretically, the elementary radiative recombination of electrons with bare nuclei, similar to bremsstrahlung, was studied when atomic quantum physics developed. Experimental studies began around 1950 and revealed that for many-electron atoms and for molecules, the binary recombination rates exceeded those of the elementary radiative process mostly by orders of magnitude. For atomic ions, this was explained by arrays of electron capture resonances, stemming from the numerous internal excitation pathways of many-electron systems (dielectronic recombination) [70]. For molecular ions, it was understood that their internal complexity opens an additional, fundamentally different energy dissipation mechanism: dissociation into neutral fragments (dissociative recombination) [71]. Theory is highly challenged by the many-particle character of the processes for both, atoms and molecules, and succeeded to develop predictive models for the less complex cases. Experiments were advanced by isolating the binary processes using ion beam storage rings and merged electron beams. There, also particle counting and imaging methods were applied to analyze the products of single recombining collisions. Still, the reliable modeling of binary electron-ion recombination is not possible for many of the collision systems relevant in plasma environments.

**Current and future challenges.** For atomic ions, storage-ring experiments [70] enabled systematic studies for individual charge states at high collision energy resolution. Recent studies questioned the habitual picture of dielectronic capture, where single-electron excitation occurs under simultaneous binding of the incident electron. Several experiments found resonances involving multiple electron excitations. Moreover, for atomic ions with complex configurations such as open f-shells, the experimental cross sections turned out to be much higher than predicted by ‘dielectronic’ recombination theory and could be approximately reproduced only by assuming



**Figure 11.** Dielectronic recombination cross-section of  $\text{W}^{18+}$  ions ( $4f^{10}$  valence configuration) measured with the storage-ring merged-beams method. (Cross section  $\sigma$  multiplied with the collision velocity  $v$  and convolved with the electron energy spread.) Calculations show radiative recombination (RR), dielectronic recombination by one-electron promotion in intermediate coupling (IC), and a model including statistical partitioning and a damping correction (PD). (Adapted from [72], copyright 2014 by the American Physical Society. Beam-merging artefacts at  $<0.001$  eV are negligible in the plasma rate constants.)

statistical multi-electron excitation (figure 11) [72]. The theory of statistical excitation is not yet sufficiently developed to replace detailed measurements of recombination rate constants for many highly charged ions, including species of interest for fusion-related plasmas. This underlines the need to further consider the roles of energy dissipation and non-reversibility that fundamentally accompany any binary recombination process.

For molecular ions, the quantum physics of capture resonances becomes even more complex through the molecular excitation paths. At low collision energy, narrow resonances occur by coupled non-adiabatic dynamics of electrons and nuclei, especially when the Rydberg orbit of a captured electron couples to rotational and vibrational motion. Moreover, initial rotations and vibrations are important for the energetic positions of these resonances. The role of rotational excitation in dissociative recombination of molecular ion turned out to be significant in recent theoretical studies [73]. But related experiments, in particular by the storage-ring merged beams technique, were so far restricted to few, infrared-inactive molecules [71]. The availability of fast ion beams in cryogenic electrostatic storage rings [74, 75] is about to change this situation.

The importance of electron-ion recombination in plasma physics and gas-phase ion chemistry leads to strong demand of reliable rate coefficients for a wide range of ionic species. Much effort has recently been spent on conversion procedures for cross section data from binary collision studies to plasma rate constants [76] and storing those in databases for interdisciplinary use. Including there the sensitivity to initial ion excitation, required for various environments, will be a future task as such dependences are becoming known.

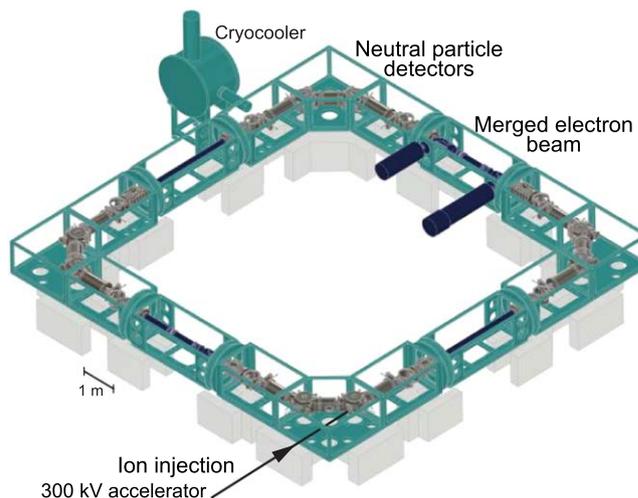
*Advances in science and technology to meet challenges.* Ion chemistry and plasma-related collisions are studied by a wide range of techniques, including discharge and flow devices, plasma reactors, electron-beam ion traps and ion beams. In some of these, the excitation temperature is well controlled by inert buffer gas. In others, energetic collisions enable very high, though typically not single, charge states. Thus, all methods offer merits and complement each other.

The best environment for in-vacuum binary collision studies is arguably offered by ion beams, in particular inside an ion storage ring. In the recently realized cryogenic electrostatic storage rings [74], ion beams of  $\sim 20$  to  $\sim 300$  keV kinetic energy can be stored for minutes to hours, while blackbody radiation is reduced to  $\sim 10$ – $20$  K effective temperatures. Stored molecular ions then can internally cool by infrared emission. A photocathode-based merged electron beam suitable for electron–ion recombination studies was recently added to one of these cryogenic rings (figure 12) [75]. It will enable to study how rotational excitation modifies the dissociative recombination of various di- and polyatomic ions with cold electrons. A detailed understanding of the rotational cooling in the stored ion beam will help to extract recombination cross sections even for specific rotational levels. These data can be expected to boost the knowledge on electron–ion recombination at the low temperatures relevant for interstellar molecular clouds.

With advanced particle imaging detectors, the merged-beams technique can also analyze the masses of the neutral products and collision kinematics of molecular electron–ion recombination. Multi-pixel cryogenic microcalorimeter detectors are under development for this purpose [77]. They will particularly benefit the study of polyatomic molecules. Aside of the large interest in branching ratio data on the neutral products, the increasing molecular complexity also carries important fundamental aspects. Thus, the possible energy dissipation into multi-mode vibrations of the products may modify the branching ratios and overall rates and even lead to non-dissociative recombination.

Reducing the operating temperature of discharge and flow-tube experiments on dissociative recombination to  $\sim 10$  K or below is a challenge actively pursued [78]. Also laser techniques are being implemented to analyze the rotational level populations of the studied molecular ions.

Electron recombination with atomic ions of complex configurations will be best addressed by merged-beam studies. High charge states of interest for fusion plasma research will require high-energy storage ring facilities, while



**Figure 12.** The cryogenic electrostatic storage ring CSR that started operation in 2015 with the recently added electron beam section. The elements enclosing and steering the ion beam, the electron collision region and the particle detectors are operated below 10 K. (Adapted from [75] with the permission of AIP Publishing.)

the new cryogenic low-energy rings will be suitable for the so far widely unexplored recombination of heavier singly and doubly charged ions.

*Concluding remarks.* As an important gas-phase ion process, electron–ion recombination in binary collisions continues to challenge theory. Thus, multi-electron excitation and, where applicable, the molecular degrees of freedom of the target and the neutral products must be explicitly addressed to successfully model the process. The ongoing development of suitable methods [73] can be critically supported by experiments, which are accessing the cold ( $\sim 10$  K) regime regarding both collision energies and molecular excitations. Moreover, also heavier atomic ions with complex electron configurations and polyatomic molecules can be addressed, where statistical energy dissipation can affect the binary collision process in yet unexplored ways. Electron–ion recombination will finally remain an important technique to populate and study atomic states in highly-charged few electron systems. Furthermore, in the regime of high collision energies where binary processes can include the interaction of fast ions with quasi-free electrons in dense targets, even electron–ion recombination mediated by transitions in the atomic nucleus is considered [79].

## 9. Low-energy electron interactions with biomolecules

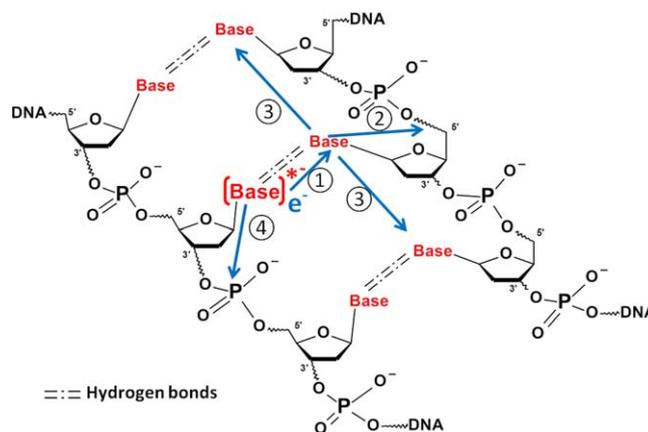
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**Status.** When high-energy radiation (HER) is absorbed into biological media, the immediate products are ions and secondary electrons. Secondary electrons are the most numerous species created by HER and their initial distribution peaks around 9–10 eV, irrespective of the mass and energy of the ionizing particle. Hence, a considerable amount of the damage to irradiated biological tissues is expected to result from the interaction of low-energy electrons (LEEs) with complex biomolecules, including those vital to life. The first observation of LEE-induced damage to such a vital biomolecule (i.e. DNA) demonstrated that, not only single and double strand breaks (SSBs and DSBs) were produced by 5–20 eV electrons, but below about 15 eV, the damage was almost completely derived from transient anions (TAs) formed on basic constituents [80]. Shortly after, TA formation in DNA was confirmed theoretically by calculations on electron attachment to a mononucleotide. This work showed preferential electron capture by the base below 5 eV, followed by electron transfer from the base to the phosphate group and decay of the TA on that group into dissociative electron attachment (DEA) [81]. DEA to bacterial DNA was later confirmed in this energy range by the experimental observation of two TAs between 0 and 4 eV, causing SSBs [82].

These initial observations were followed by numerous experimental and theoretical studies on various molecules, ranging from gaseous or condensed amino acids, peptides, DNA basic constituents (bases, sugar and phosphate group), to condensed oligonucleotides, origami oligonucleotide arrays, self-assembled monolayers of short DNA strands and bacterial (i.e. plasmid) DNA [82–84]. The condensed-phase targets usually consisted of multi- or sub-monolayer films of the biomolecule deposited on a conductive substrate. The type of damages in DNA, analyzed with various techniques, included bond cleavage, SSBs, base damage, DSBs and more complex clustered lesions, consisting of SSBs and base damages. From yields versus energy functions, it was generally concluded that the decay of TAs into destructive channels played a major role in inducing the lesions. These channels include DEA and production of dissociative electronic neural states resulting from TA autoionization. However, in DNA, electron transfer between basic units increases the complexity of the damage mechanism. The probable pathways leading to various DNA lesions [85] are summarized in figure 13.

Defining the role of LEEs in radiobiology constitutes an essential element of our understanding of the action of HER on living organisms, including humans. Such comprehension has implications in radiotherapy alone, or combined with chemotherapy [86, 87], as well in other applied areas, including estimations of the risks of health effects from HER



**Figure 13.** Within DNA, a TA is usually formed on a base and then decays into DEA or autoionization. In the latter case, the electron released can transfer to other subunits via pathways 1–4 to produce a TA at another site, accompanied or not by base dissociation. Depending on the pathway, the DNA lesions mentioned in the text can be produced. Reprinted with permission from [85]. Copyright 2017 American Chemical Society.

present in the environment on earth and in space. In these cases, the dose is low, and hence, knowledge of the details of energy deposition at the micro- and nano-scopic level within cells may be crucial to evaluate the biological risks. Such details are also needed in the development of new radiotherapeutic modalities, including targeted radionuclide therapy (TRT) and nanoparticle-aided and heavy-ion radiotherapy [86, 87]. In these emerging cancer treatments, high local biological effectiveness and preferential reduction of cancer cells is achieved by producing large localized densities of LEEs, which decrease radiation damage in healthy tissues [86, 87]. Moreover, since many radiosensitizers and chemotherapeutic drugs amplify LEE-induced damage, fundamental knowledge of LEE-interactions is expected to improve treatments with concomitant chemoradiation therapy (CRT) [87]. Recent pre-clinical studies have shown that basic information on the action of LEEs on Pt-chemotherapeutic drugs bound to DNA could predict the time delay between intravenous administration of the drug and tumor irradiation, necessary to obtain the most efficient CRT [88]. Besides HER-related applications, experiments on gaseous biomolecules, with the support of *in silico* methods, have served to correlate the activity of various classes of biological compounds with fragment species formed by DEA [89].

**Current and future challenges.** Biological effects of LEEs and the secondary species created by them or by the parent ions (e.g. OH and H radicals and solvated electrons) have been investigated in models environments. Ideally, the interaction of these species and LEEs with biomolecules should be investigated within cells, where the configuration of a given biomolecule is influenced by the presence of not only water, but also by that of ions, proteins and other biomolecules. LEE experiments on biological DNA were performed with thin films in vacuum and under atmospheric conditions in the presence of small cellular constituents, such

as  $O_2$  and water. Thus, the ultimate challenge for the field consists in obtaining a complete knowledge of the mechanisms leading to LEE-induced damage within living cells. It is, however, difficult to imagine how this is possible theoretically and experimentally. Even if one could measure the damage created by monochromatic LEEs injected at a certain position in a cell, without perturbing its functions, how could the reaction pathways be determined? At least for the next decade, it is probably more realistic to assemble knowledge from various biologically relevant experiments and calculations, to achieve an accurate vision of the biological action of LEEs.

There are at least five main avenues of future developments, presenting considerable experimental and theoretical challenges: (1) refinement of thin film fabrication on surfaces, (2) development of technology capable of investigating the reactions of LEEs with biomolecules in solution, (3) refinements of cluster beam technology to progressively embed in a given medium more and larger biomolecules, (4) enhancement of theoretical and computational capabilities to treat more complex and larger LEE-biomolecule systems, so as to incorporate the effects of solvation, bimolecular environment and reactions of initial products and (5) refinement of Monte-Carlo codes to include adequately LEE scattering and attachment cross sections and thus provide reasonable estimates of the local dose distributions and biological effectiveness at the level of the cell, nucleus and DNA.

*Advances to meet these challenges.* Multi- or sub-monolayer film experiments in vacuum have the advantage of a monochromatic electron source, whose energy can be varied from close to zero to hundreds of eVs. To emulate cellular conditions, new nanoscale techniques need to be developed to assemble biomolecules in particular configurations on a substrate. Many of the cellular structures and properties of molecules may thus be reproduced. Furthermore, the configurations and types of molecules could be modified to extract the behavior of different environments. Ideally, one should be able to interrogate directly,

at specific positions in the film, damage and formation of new complexes. The origami-template approach is a good example showing that DNA strands can be constructed in a specific configuration on a substrate and the damage can be measured *in situ* [84]. More generally, continued advances in both thin film and biomolecular-cluster technology would benefit from the development of more sophisticated methods to evaporate large biomolecules in vacuum, including further elaboration of laser-induced desorption. It is also becoming possible to investigate LEE-biomolecule interactions in water or other biologically relevant liquids, by secondary- or photo-electron emission from immersed surfaces, ultra-fast pulse radiolysis and femtosecond laser-induced low density plasma. Depending on the energy of the incident photons, the first method can generate electrons ranging from a few to hundreds of eVs. Pulse radiolysis can time-resolve intermediate species, whereas the latter method generates only LEEs below 8 eV. Future progress relies on increased knowledge of the chemistry introduced by scavengers, particularly their reaction with LEEs. Such experiments could be supported by realistic theoretical models. Moreover, quantum mechanical calculations could be performed on a combination of a chemotherapeutic drug, a radiosensitizer or nanoparticle bound to a target molecule (e.g. DNA) to estimate the enhancement of LEE-induced damage and learn more about the mechanisms of radiosensitization.

*Conclusion.* Secondary electrons possess most of the energy injected into biological tissue by HER. Advances in LEE-biomolecules interactions have shown that a considerable portion of this energy flow can be redirected efficiently, thus enabling the production of more potent radiosensitizers and the development of improved clinical protocols. Considering the value of such advances for radiotherapy, CRT and health hazards estimates of HER, the field is expected to grow appreciably in the next decades. More generally, information on electron interaction and attachment to biomolecules is likely to unveil initial molecular mechanisms of electron-driven biological processes.

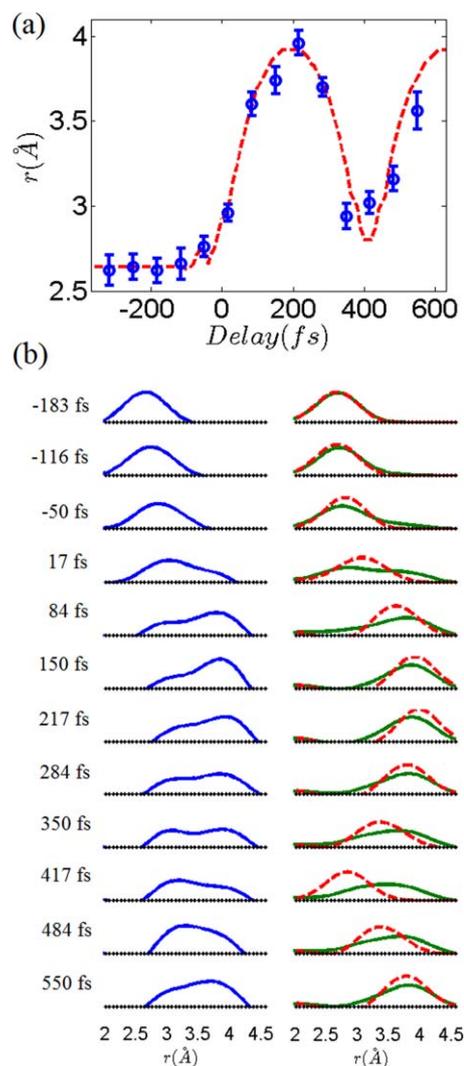
## 10. Ultrafast electron diffraction from photo-excited molecules in the gas phase

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**Status.** The conversion of light into mechanical and chemical energy at the single molecule level takes place through the motion of electrons and nuclei. Following light absorption, changes in electronic potential occurring on the attosecond timescale lead to the motion of nuclei towards a new minimum on the potential energy surface, typically on the femtosecond timescale. This simple picture, however, breaks down when the nuclear and electronic motions are coupled, for example when the nuclear wavepacket approaches a conical intersection. The development of spectroscopic probing methods based on the femtosecond laser gave rise to the field of femtochemistry, which enabled the measurement of coherent molecular dynamics in real time. Most spectroscopic methods probe changes in the energy landscape of the electrons that interact with the laser light, however, these methods are not directly sensitive to the motion of the nuclei. Gas electron diffraction (GED) has been used to determine the structure of isolated molecules with very high precision [90]. The introduction of femtosecond laser pulses to trigger the emission of short electron pulses gave rise to the field of ultrafast electron diffraction (UED), where a laser pulse is used to start a photochemical reaction and an electron pulse is used to probe the changes in molecular structure. Although UED experiments with picosecond resolution were used to determine the structure of short-lived species [91] and to capture three-dimensional images of laser-aligned molecules [92], these lacked the temporal resolution required to observe the coherent nuclear motion that takes place on femtosecond timescale.

The temporal resolution of UED experiments had been limited by the Coulomb broadening of electron pulses and the temporal smearing associated with having laser and electron pulses traverse the sample at different velocities. Through the use of compact electron guns which minimize the propagation distance of the electron pulses, and samples with thickness below 100 nm, condensed matter UED overcame these limitations to reach femtosecond resolution [93]. In gas phase UED, femtosecond resolution was achieved through the use of relativistic electron pulses, which minimize the detrimental effects of both the Coulomb broadening and the velocity mismatch [94]. The use of relativistic electrons led to the first observation of coherent nuclear motion in a gas phase UED experiment [95]. In this experiment, iodine molecules were excited to a bound electronic state using a femtosecond laser pulse, inducing a large amplitude coherent vibration. This motion was captured by UED with 220 fs resolution (figure 14(a)), and the changing bond length was retrieved with a precision of 0.07 Å. Most importantly, this experiment demonstrated UED's ability to not only retrieve the interatomic distances, but also to capture the shape of the moving nuclear wavepacket, as shown in figure 14(b).



**Figure 14.** (a) Time-resolved experimental (blue circles with error bars) and simulated (red dashed line) bond lengths retrieved from the UED data. (b) The experimental (left panel) and simulated (right panel) time-resolved probability density function of the retrieved nuclear wavepacket. The dashed red lines are the results of the theoretical calculations with the same spatial resolution as the experiment, and the solid green lines include also averaging due to the temporal resolution (230 fs). The black dotted line indicates the baseline for each curve. Reproduced from [95].

Recent experiments with an improved temporal resolution of 150 fs have targeted more complex systems, where the capability of UED to directly capture the nuclear motion is critical for revealing the structural dynamics. In Trifluoriodomethane ( $\text{CF}_3\text{I}$ ) molecules excited with a UV femtosecond laser pulse, UED has revealed the splitting of the nuclear wavepacket as it passes through a conical intersection (where two potential energy surfaces meet) [96]. Following excitation, a repulsive force between the carbon and iodine atoms causes the bond length to increase. When the system reaches the conical intersection, the nuclear wavepacket splits, allowing part of the population to return towards the initial bond distance. This results in a coherent oscillation where some of the population escapes towards dissociation at each

turn. The dynamics were retrieved by analyzing the angularly resolved Fourier Transforms of the diffraction patterns.

*Current and future challenges.* There are multiple systems and important reaction types that can be investigated with the current UED capabilities. The study of model systems, such as the ring-opening reaction of Cyclohexadiene, cis-trans isomerization of Stilbene and Azobenzene and relaxation dynamics in the nucleobases, are currently being pursued by UED. However, further improvements in temporal and spatial resolution, structural retrieval algorithms, and sample preparation and delivery are required in order to investigate more complex systems and systems undergoing faster and/or more subtle structural changes.

There are dynamical processes, for example proton transfer and roaming reactions, which require a temporal resolution on the order of 20 fs. Detecting small structural changes, such as the motion of a single light atom, will require improvements in both spatial resolution and signal to noise ratio. Currently UED captures a range of momentum transfers of up to approximately  $12 \text{ \AA}^{-1}$ , being limited by the flux of scattered electrons. An improvement of a factor of two in the captured momentum transfer (scattering angle) would place UED on par with static GED and allow for retrieving distances with comparably high accuracy.

Another current limitation is the lack of a general algorithm for retrieving structures from the diffraction patterns. Structure retrieval methods routinely used in static GED, have been found to be unsuitable in the context of UED, as the sample volume contains both unexcited and excited molecules, which may follow multiple different paths to the final product, or products, and exhibit large amplitude vibrations.

Finally, the high sample density required to achieve a sufficient flux of scattered electrons represents a challenge in sample preparation and delivery. In some cases, the current method of heating a sample to increase the vapor pressure to the order of tens of mBars has been found to be unsuitable, as molecules may thermally decompose before the desired vapor pressure is reached.

#### *Advances in science and technology to meet challenges.*

The temporal resolution in UED experiments is determined by the duration of the laser pulse that excites the sample, the duration of the electron pulses, the velocity mismatch between laser and electron pulses, and the synchronization between the laser and electron pulses. Laser pulses of less than 20 fs can be routinely produced, while the velocity mismatch becomes insignificant for the electron energies of a few MeV. The remaining challenges are minimizing the electron pulse duration and the timing jitter between laser and electron pulses.

The duration of electron pulses can be reduced using an RF buncher cavity [97]. It was recently shown that an MeV electron

pulse could be compressed to a duration of less than 10 fs [98]. However, RF compressors introduce a timing jitter, typically in the order of hundreds of femtoseconds, which limits the temporal resolution. Correcting the timing jitter at the 10 fs level will require the use of a time stamping technique to measure the time of arrival of each electron pulse with respect to the laser pulse. This could be achieved by deflecting the electron beam with a THz laser pulse or streak camera, or using the interaction of the pulses with a material. Time stamping requires that the scattering pattern of electrons be recorded for each electron pulse separately, which will require improvements in detector technology in order to run experiments at high repetition rates. A detector with fast readout that can operate in electron counting mode would significantly improve the signal to noise ratio of the experiments by removing noise from individual frames before averaging.

The use of high repetition rate electron guns will undoubtedly improve the spatial resolution of UED experiments, particularly when working with lower density samples. Pulsed photo-electron guns with relativistic energy and MHz repetition rates have been demonstrated [99]. This represented a four-order of magnitude improvement compared to the existing 120 Hz relativistic guns in operation. Implementing this high repetition rate guns into a UED setup would also require a high repetition rate laser with a sufficiently high pulse energy to excite the sample and drive the electron gun. The average power of such laser system is expected to exceed a kW.

New structure retrieval algorithms are needed to tackle the challenges unique to UED, i.e. multiple transient structures that can become partially delocalized due to wavepacket dispersion and large amplitude vibrations. Finally, the challenge of delivering large molecules onto the target volume, at high density, still remains unresolved.

*Concluding remarks.* Recent improvements in technology have allowed UED to image coherent nuclear motion in photo excited molecules with atomic resolution. Recent and ongoing experiments are providing spatially resolved measurements of previously inaccessible aspects of reaction dynamics such as the splitting of nuclear wavepackets and coherent structural dynamics. UED is, in principle, applicable to more complex systems than those studied so far, and with further technological improvements could capture even the fastest nuclear motions with high spatial resolution.

## Acknowledgments

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## POSITRONS AND ANTIMATTER

### 11. Atomic collision processes to synthesize antihydrogen and exotic atoms for anti-matter research

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*Status.* At the beginning of the universe, the Big Bang, ca 13.8 billion years ago, the same amount of matter and antimatter are predicted to be born via pair production processes according to the best knowledge of the present elementary particle physics. During the expansion immediately after the Big Bang, most of matter and antimatter vanished via reverse processes, i.e. pair annihilation. A natural consequence of this scenario is that antimatter world should exist some-where in the universe having exactly the same amount of (anti-)material as our matter world. However, no astronomical observation gives any slight signs of antimatter world strongly indicating that the production and/or annihilation processes are asymmetric between matter and antimatter in some way. Consequently, our matter world luckily survived keeping enough material, which gives us a chance to develop as thinking reed and now we are wondering why and how we survived.

One of the key concepts to attack this missing antimatter mystery is the fundamental symmetry, charge, parity, and time reversal (CPT) symmetry. Actually, the CPT symmetry is derived from the Standard Model constructed on a flat space and time, and leads to identical spectra in matter and antimatter. Because of this, the comparison of antihydrogen ( $\bar{\text{H}}$ ), the unique antimatter stable and accessible, with hydrogen (H) has a high potential to test the CPT symmetry although some precision tests were done with other systems like  $\bar{\text{K}}^0$  and  $\text{K}^0$ .

Another important subject with antimatter is to study the weak equivalence principle (WEP), which has been tested to very high precision for a range of material compositions. However, no such precision test has ever been done between matter and antimatter. It would then be highly attractive to make such a test using the Earth and  $\bar{\text{H}}$  atoms as the experimental system.

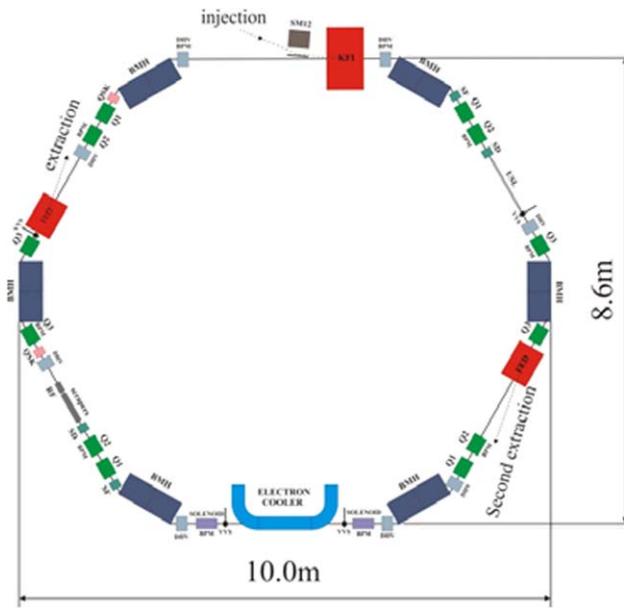
Regarding the antihydrogen research, the first  $\bar{\text{H}}$  production was in 1996 by Oelert and his colleagues who synthesized fast  $\bar{\text{H}}$  atoms shooting 1.4 GeV antiprotons ( $\bar{\text{p}}$ s) on Xe atoms. The involved collision mechanism is a charge transfer process like  $\bar{\text{p}} + \text{Xe} \rightarrow \bar{\text{H}}(\bar{\text{p}}\text{e}^+) + \text{e}^- + \text{Xe}$ , i.e. a positron capture from the Dirac's 'positron sea' leaving a positron hole, i.e. an electron, in vacuum [100]. In 2002, the ATHENA collaboration successfully synthesized 'cold'  $\bar{\text{H}}$  atoms in highly-excited states employing a three-body recombination collision like  $\bar{\text{p}} + \text{e}^+ + \text{e}^+ \rightarrow \bar{\text{H}}^* + \text{e}^+$  [101]. The ATRAP collaboration followed immediately using the same scheme in the same year. Two years later, the ATRAP collaboration successfully demonstrated a new scheme adopting three successive atomic collisions like  $\text{Cs} + h\nu \rightarrow \text{Cs}^*$ ,  $\text{e}^+ + \text{Cs}^* \rightarrow \text{Ps}^*(\text{e}^+\text{e}^-) + \text{Cs}^+$ ,  $\bar{\text{p}} + \text{Ps}^* \rightarrow \bar{\text{H}}^* + \text{e}^-$  [102]. In 2010, the ALPHA collaboration succeeded in trapping  $\bar{\text{H}}$  atoms in a magnetic bottle [103], and the ASACUSA

collaboration made a beam of cold  $\bar{\text{H}}$  atoms [104], both of which are the first step to physics research using  $\bar{\text{H}}$  atoms. In 2018, the ALPHA collaboration eventually succeeded to measure 1S–2S transition of  $\bar{\text{H}}$  atoms with the relative precision of  $2 \times 10^{-12}$  confirming the CPT symmetry at this level regarding the 1S–2S transition [105]. In 2017, the BASE collaboration determined the magnetic moment of  $\bar{\text{p}}$  with better precision than that of p. This was an epoch-making event in the sense that the property of an exotic particle nonexistent in nature was known better than that of the most abundant particle in our universe, the proton [106]. Although the antihydrogen research progressed a lot in the last two decades as quickly described above, there are still a lot to improve to really attack the CPT symmetry and the WEP.

The AEGIS collaboration aims at synthesizing a pulsed beam of cold  $\bar{\text{H}}$  atoms in specific quantum states by showering a pulsed positronium ( $\text{Ps}^*$ ) cloud in excited states on trapped cold  $\bar{\text{p}}$ s like  $\text{Ps} + h\nu \rightarrow \text{Ps}^*$ ,  $\bar{\text{p}} + \text{Ps}^* \rightarrow \bar{\text{H}} + \text{e}^-$ .  $\text{Ps}^*$  has much longer lifetime and at the same time has much higher antihydrogen formation cross sections than Ps in its ground state, which are both essential to effectively produce  $\bar{\text{H}}$ . Then,  $\bar{\text{H}}$  atoms in highly excited states are manipulated employing a Stark acceleration, which yields a well-controlled  $\bar{\text{H}}$  beam. AEGIS is going to measure the gravitational force on such a  $\bar{\text{H}}$  beam using a so-called Moire deflectometer. The Gbar collaboration aims to make  $\bar{\text{H}}^+$  ( $\bar{\text{p}}\text{e}^+\text{e}^+$ ) ions, the antimatter of  $\text{H}^-$  ( $\text{p}\text{e}^-\text{e}^-$ ) via two successive charge transfer collisions shooting  $\bar{\text{p}}$ s in a high-density  $\text{Ps}^*$  cloud like  $\bar{\text{p}} + \text{Ps}^* \rightarrow \bar{\text{H}} + \text{e}^-$ ,  $\bar{\text{H}} + \text{Ps}^* \rightarrow \bar{\text{H}}^+ + \text{e}^-$ . Because the  $\bar{\text{H}}^+$  ion is positively charged, it can be sympathetically cooled by, e.g. laser-cooled  $\text{Be}^+$  ions as low as a few nano eV. By photo-detaching one positron from cooled  $\bar{\text{H}}^+$ , free fall measurements with ultracold  $\bar{\text{H}}$  are going to be realized. It would be interesting and important to figure out other atomic collision processes to produce  $\bar{\text{H}}^+$ s efficiently. The ALPHA collaboration is also planning to make a free fall experiment laser-cooling the trapped  $\bar{\text{H}}$ s.

*Current and future challenges.* Additional tests of symmetry and WEP can be realized employing matter–antimatter systems consisting purely of baryonic or leptonic particles like protonium ( $\bar{\text{p}}\text{p}$ ) and Ps. Although they eventually annihilate, the lifetime can be elongated drastically by exciting such systems to higher ( $n$ ,  $l$ ) states. It is noted that  $\bar{\text{p}}\text{p}$  and  $\bar{\text{p}}\text{A}^{q+}$  formation processes provide interesting atomic collision system with  $\bar{\text{p}}$  as a 'heavy electron'. Further, the annihilation pattern of  $\bar{\text{p}}\text{A}^{q+}$  would give information on the 'surface structure' of the nucleus, which is particularly interesting for unstable nuclei (PUMA collaboration).

Any particle in a sufficiently deep and wide potential well can be bound in discrete quantum states, which had been demonstrated for ultra-cold neutrons in the gravitational field falling against a horizontal mirror [107]. The Gbar collaboration is planning to make such experiments using their ultra-cold  $\bar{\text{H}}$  atoms. The falling  $\bar{\text{H}}$  atoms do not move continuously along the vertical direction, but rather jump from one height to another.



**Figure 15.** A schematic drawing of ELENA under commissioning. Pulsed 5.3 MeV  $\bar{p}$ s from the AD are injected from the upper part ('injection'), cooled by the electron cooler and decelerated down to 100 keV, and finally transported to each experiment. By courtesy of CERN (<https://espace.cern.ch/elena-project/SitePages/Home.aspx>).

The sidereal variation which was studied for the charge to mass ratios of  $p$  and  $\bar{p}$  by the BASE collaboration in 2015 would be worth tested further with other physical quantities.

An interesting future subject would be to replace  $e^-/e^+$  in  $H/\bar{H}$  by  $\mu^-/\mu^+$  and make high-precision spectroscopy of muonic hydrogen ( $\mu^-p$ ) and muonic antihydrogen ( $\mu^+\bar{p}$ ). Such an experiment could be realized by transporting trapped  $\bar{p}$ s to a muon facility or constructing a compact muon facility next to the antiproton decelerator (AD) at CERN. Because  $\mu^-/\mu^+$  is  $\sim 200$  times more massive than  $e^-/e^+$ ,  $\mu^-p/\mu^+\bar{p}$  is  $\sim 200$  times smaller than  $H/\bar{H}$ , and so this system could probe the properties of  $p$  and  $\bar{p}$  with much higher sensitivity. In 2010,  $\mu^-p$  was synthesized by injecting low energy  $\mu^-$  beam in a  $H_2$  gas cell, and its  $2s-2p$  transition was measured. It was concluded that the resulting proton radius is smaller than that evaluated by high energy electron scattering experiments, which was a big news in the related community [108]. Regarding the  $\mu^+\bar{p}$  formation, it would be realized e.g. by a charge exchange collision like  $\bar{p} + \mu^+e^- \rightarrow \mu^+\bar{p} + e^-$ , where  $\mu^+e^-$  is reasonably produced by injecting slow  $\mu^+$  in various solid material like silica aerogel.

A groundbreaking new era of the cold antimatter research would start if antideuteron ( $\bar{d}$ ) is available, which enables high precision antimatter research involving antineutron ( $\bar{n}$ ) for the first time. The way how to produce  $\bar{d}$  is discussed in the next section.

*Advances in science and technology to meet challenges.* To further push the antihydrogen research, one of the most critical issues is to increase the number of usable antihydrogen atoms. In

parallel to various efforts to increase the production efficiency, a new decelerator ring, the Extra Low ENergy Antiproton ring (ELENA), was constructed at CERN to increase the number of  $\bar{p}$ s usable for experiments. ELENA cools and decelerates 5.3 MeV pulsed beams of  $\bar{p}$  from the AD down to 100 keV. The number of usable  $\bar{p}$ s for  $\bar{H}$  experiments is expected to be  $\sim 2 \times 10^7$  per AD shot, a factor of 10–100 more than the present. The commissioning of ELENA had already started and is expected to be ready in 2021 after LS2 (long shut down of the CERN accelerator complex starting from the end of 2018).

The temperature of a  $\bar{p}$  cloud primarily determines the  $\bar{H}$  temperature which is the key parameter for almost all the ongoing antihydrogen experiments. At the start of the cold  $\bar{p}$  research in 1986, Gabrielse and his colleagues co-trapped  $\bar{p}$ s with  $e^-$ s in a superconducting solenoid. Electrons in a strong magnetic field are automatically cooled via synchrotron radiation down to an environmental temperature and sympathetically cool  $\bar{p}$ s. With this technique,  $\bar{p}$  clouds are quickly cooled down to several tens K but not lower due to heating by various noise sources. To achieve lower temperature, a so-called evaporative cooling was developed by the ALPHA collaboration, and the temperature can be lowered by a factor of ten or so. To push further, a sympathetic cooling of an antiproton by a laser cooled  $Be^+$  ion is in progress by the BASE collaboration. To avoid annihilation between  $\bar{p}$  and  $Be^+$  they should be spatially separated but at the same time the electrical coupling should be strong enough so that  $\bar{p}$  can be efficiently cooled. It is quite important to extend this technique to a large number of  $\bar{p}$ s for ultra-cold  $\bar{H}$  research.

An elegant scheme to produce  $\bar{d}$  had already been discussed in 1982 employing a mirror process of a well-known nuclear reaction,  $p + p \rightarrow d + \pi^+$ , i.e.  $\bar{p} + \bar{p} \rightarrow \bar{d} + \pi^-$ . Dieter Möhl and his colleagues proposed to circulate two  $\bar{p}$  clouds with different momenta in a double storage ring so that they collide at the merging point [109]. If the momenta of two  $\bar{p}$  clouds are selected so that the resulting  $\bar{d}$  momentum matches with one of  $\bar{p}$  cloud momentum, the resulting  $\bar{d}$ s are automatically accumulated in one of the rings.

*Concluding remarks.* Cold antimatter research until now and in the future is briefly reviewed paying attention to the roles played by atomic collision processes. It is noted that high precision research including cold antimatter would be complementary to high energy physics to study fundamental laws of nature. This approach would get more important in a future because the accelerator size and so the particle energy will reach their limit sooner or later unless a new type of compact accelerator employing, e.g. a shorter wave length than microwave is invented.

## Acknowledgments

YY would like to thank CERN, specifically the AD team, the Radio Frequency (RF) team, and the ELENA team for their great efforts and support to the cold antimatter community.

## 12. Positron and positronium scattering experiments

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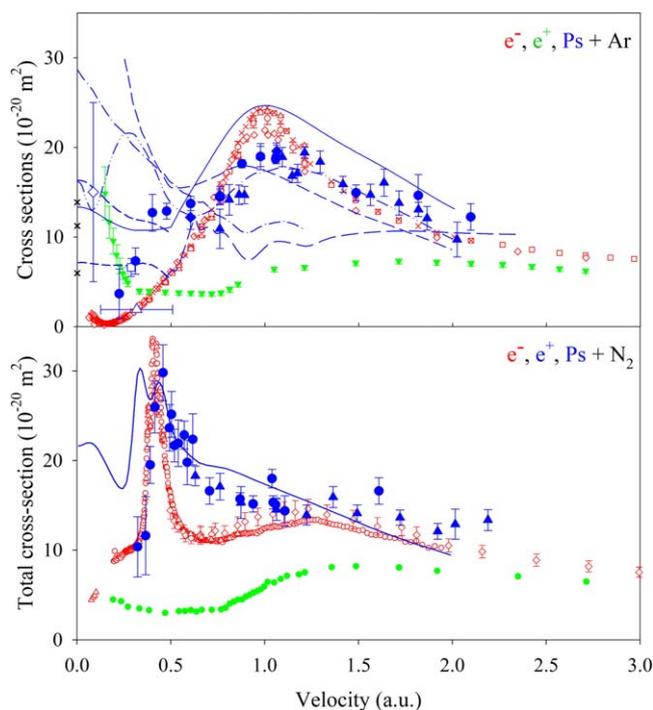
**Status.** Studies of positron ( $e^+$ ) and positronium (Ps) collisions with atoms and molecules are driven primarily by the desire to advance knowledge of basic matter–antimatter interactions as well as of atomic and molecular collision physics in general (e.g. [110–115] and references therein), the relevance of these data to other fields (as in modelling propagation and diffusion for dosimetry in positron emission tomography and astrophysical and atmospheric events) adding further incentive to this pursuit.

Over the years, information has been acquired through experimental and theoretical determinations principally of total and partial integral cross-sections but also some differential studies. This section focuses on aspects of what has been learnt, in conjunction with theory, mainly from measurements performed in the presence of relatively weak or no magnetic fields, Cliff Surko and James Sullivan in this article reviewing experiments performed with positron beams injected from electromagnetic traps.

**Positrons.** It has been known for some time that the static and polarization interactions have opposite signs and partially cancel for  $e^+$  whilst they are both attractive and add up for electrons ( $e^-$ ) [110, 112]. As a consequence, despite the presence of extra channels (i.e. annihilation and Ps formation), the positron total cross-section is usually smaller than that for  $e^-$  at intermediate energies, usually converging at high energies as illustrated in figure 16 for Ar and N<sub>2</sub> [116–119].

The total ionization cross-section for the inert atoms tends to be greater for the positively charged projectile at intermediate energies, not just because of Ps formation but also because the peak of the direct ionization cross-section may surpass that for electron impact due to polarization effects [111]. The probability of Ps formation is high with a greater propensity for forward emission from targets with a low  $Z$  (e.g. He and H<sub>2</sub>). For the inert atoms, its formation in an excited state (Ps\*) increases in going from He to Xe while little Ps\* has been observed from N<sub>2</sub> and CO<sub>2</sub> where instead Ps formation accompanied by electronic excitation of the ion is dominant and attributed to the coupling between the quasi-degenerate channels for the excitation of the neutral target and that of the ion produced via Ps formation [111]. A considerable body of work is also available for energy-resolved positron–molecule annihilation arising from positron binding and vibrational Feshbach resonances [120].

Differential studies of positron impact ionization remain in short supply [111, 113]. Comparative studies of doubly differential cross-sections by  $e^-$  and  $e^+$  have shown that the sum of the intensities of the scattered  $e^+$  and ejected electron is similar to that of the two final state electrons in the case of  $e^-$  impact. However, whilst the projectile and the ejected



**Figure 16.** Electron (red), positron (green) and Ps (blue) scattering from Ar and N<sub>2</sub>. Data points: experiments. Lines: Ps theories. For Ps: (solid symbols) total cross-section; (hollow symbols) momentum transfer cross-section. References in [116–119].

electron are indistinguishable in the latter case because of exchange, the identification is easy for  $e^+$  impact, the ejected  $e^-$  and scattered projectile populating opposite ends of the energy spectrum. Final state electron–positron correlations have been probed in triply differential measurements performed close to 0°, exposing the electron-capture to a continuum state (ECC) of Ps. Investigations have also been extended to studies of binary and recoil interactions by positron and electron impact [111, 113].

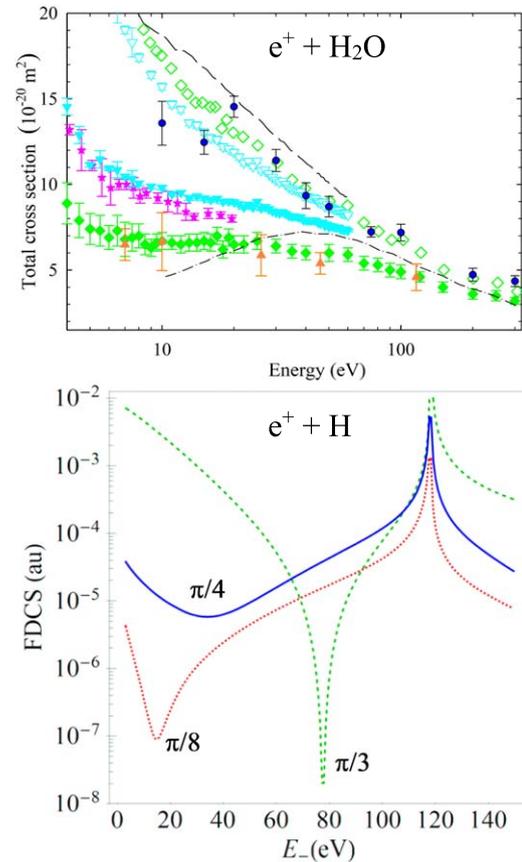
**Positronium.** Despite being neutral and twice the mass, positronium was unexpectedly found to scatter from a given target with a total cross-section close to that of an electron at the same velocity (see examples in figure 16) [e.g. 114, 116, 117]. This tendency persists in the experimental data even in the vicinity of delicate quantum mechanical phenomena occurring for low-energy electrons, such as Ramsauer–Townsend minima and resonances, as illustrated in the figure. There it can also be seen that this behaviour has stimulated considerable theoretical developments with convergence having been broadly achieved above the Ps break-up threshold ( $v = 0.5$  a.u.) while significant discrepancies remain below it [114, 116–119] in the case of Ar, while for N<sub>2</sub> a resonance is also seen for Ps both in experimental and theoretical results close to that for electrons. At low energies, the figure includes the momentum transfer cross-sections from swarm-type experiments [e.g. 114, 121, 122].

Positronium-induced ionization has been investigated in collision with He and Xe [114]. Both integral and differential cross-sections have been measured. Ps fragmentation dominates at low energies, the residual  $e^-$  and  $e^+$  remaining highly correlated in the final state, in a process known as electron-loss-to-the-continuum [114].

Ps can itself capture an additional electron to form the positronium negative ion ( $\text{Ps}^-$ ), a model system for testing three-body problems in quantum mechanics. Recently, a laser spectroscopy study of  $\text{Ps}^-$  has observed a strong shape resonance of  $^1\text{P}^o$  symmetry near the Ps ( $n=2$ ) formation threshold, the resonance energy and width found to agree with theory [123].

**Current and future challenges.** For both  $e^+$  and Ps scattering, a major focus is the search and understanding of quantum phenomena such as interference effects, resonances and diffraction. Although much data are available for the total cross-sections of positrons scattering from atoms and molecules, the variation in their energy and angular resolutions is significant, and particularly problematic for polar and highly polarizable targets where forward scattering effects are critical, as illustrated in figure 17 for  $\text{H}_2\text{O}$  [124]. Some discrepancies remain also for the integral cross sections for Ps formation and direct ionization [110–113]. More sparse are data for elastic scattering as well as molecular and electronic excitation [110, 112]. Higher angular and energy resolution (as well as intensities) are needed to quantify and control systematic effects in measuring total and elastic scattering cross-sections, also by probing differential elastic scattering at angles  $\leq 15^\circ$ . The examination of the threshold behaviour of inelastic processes and their interference with other (elastic and inelastic) channels remains topical [125]. The exploration of the cause of the anomalous yields in the angular emission of positronium (e.g. from  $\text{H}_2$  and  $\text{O}_2$ ) might clarify the influence from accidental resonances [111]. The predicted occurrence of quantum vortices in positron-impact ionization [126] requires doubly- or triply-differential studies over a range of emission angles. Measurements at very low (down to thermal) energies and the study of bound states of positrons and electrons (including precision measurements of binding energies and resonances) would fill an important knowledge gap in the interactions of  $e^+$  and Ps with matter. In the case of Ps, investigation of scattering at high energies might be facilitated by a beam produced via photodetachment of  $\text{Ps}^-$  [114, 123] whilst recently developed control techniques [127] might enable investigations at very low energies.

Recently, the lognormal function, widely used for the probability distribution of *macroscopic* stochastic events, has been found to describe also the energy dependence of inelastic collisions (including ionization, electron capture and excitation by electrons, positrons, protons, antiprotons, etc), simply by allowing for the relevant threshold energy [115]. The analysis is very general, and observed to extend also to nuclear reactions and to solid state problems. As well as



**Figure 17.** Top: total cross-sections for  $e^+ + \text{H}_2\text{O}$ : direct measurements (solid symbols), corrected for forward scattering effects (hollow symbols) and theories (lines), see [124]. Bottom: triply-differential ionization cross-section for  $e^+ + \text{H}$  at 250 eV as a function of the  $e^-$  energy ( $E_-$ ) ejected at various angles. The peak at around 120 eV signals ECC whilst the deep minimum near 80 eV at  $\pi/3$  corresponds to a quantum vortex [126] (© IOP Publishing Ltd. CC BY 3.0).

aiding the description of collision probabilities for quantum systems, this finding raises interesting questions on the fundamental issue of the interface between the classical and quantum domains.

#### Advances in science and technology to meet challenges.

The major difficulty in advancing positron and positronium experimental studies to the level reached with electron measurements is simply their beam intensities being lower by a billion or more. However, since the early days of positron physics, much ingenuity has been employed to circumvent (at least in part) this severe hindrance. Very low background levels, long-term apparatus stability and coincidences techniques are crucial in enhancing signal-to-background levels and in isolating specific processes. Yet as described above, many questions remain.

A major technical advance in positron physics has been the accumulation of positrons in a trap feeding a magnetic beamline, as described by Cliff Surko in this article. If current efforts towards coupling such device to an

electrostatic system would succeed in delivering higher intensity beams with a small spot size ( $\leq 1$  mm) and low angular divergence ( $\leq 1^\circ$ ) in a field-free interaction region, it would represent a key development for meeting some of the above challenges.

*Concluding remarks.* Experimental and theoretical studies with positron and positronium are exacting but the rewards of dealing with light quantum particles of positive or zero charge

continue to be valuable in gaining glimpses of the workings of nature at a very fundamental level.

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### 13. Physics with positron traps and trap-based beams: overview and a look to the future

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**Overview and status.** Positrons are important in many areas of science and technology. Examples include positron emission tomography (PET) which is a workhorse in medicine and drug design; numerous positron-based techniques used to study materials and material surfaces; tests of fundamental physics; and positron interactions in astrophysical settings ranging from the electron–positron plasma at the poles of neutron stars to positron annihilation in the interstellar medium.

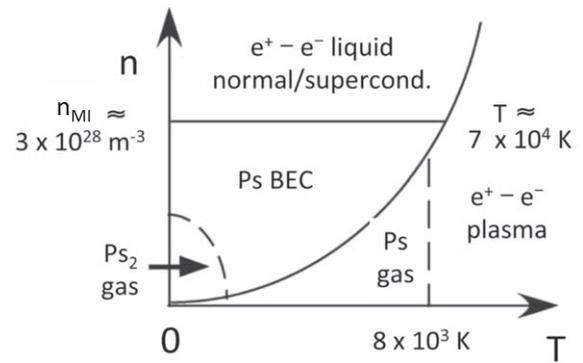
Many important aspects of physics with positrons come from experiments involving atomic and molecular physics [110]. However, a key impediment to these and other studies is the fact that sources of antiparticles in our world of matter are relatively weak as compared to their matter counterparts (e.g. 1 A to 1 kA electron beams as compared to 1 pA positron beams). Thus, it has proven advantageous, and often necessary, to develop new methods to use antimatter more efficiently.

The method of choice for many applications is to accumulate antiparticles in an electromagnetic trap in vacuum, tailor the resulting charge cloud, and then tailor the method of delivery with a specific application in mind [128]. Particular studies might require intense bursts of antiparticles, bursts delivered in short times (e.g. <1 ns), very cold antiparticles, or tunable, high-energy-resolution beams. Science in several areas has benefited from recent developments in this technology.

**Antihydrogen studies.** Positron and antiproton plasmas have been created, cooled and manipulated to form and study antihydrogen. This has enabled, for example, the first measurements of the 1S–2S transition in antihydrogen [129] and measurement of its magnetic moment [128]. Tests of the gravitational interaction between antihydrogen atoms and our (matter) earth are in progress [128].

**Physics with positronium.** Positronium (Ps) is the bound state of a positron and an electron. The generation of intense bursts of positrons has opened the door for new Ps-laser-physics [127]. For example, the creation of long-lived, high-Rydberg states of Ps and Ps Stark states (i.e. formed in an electric field) has led to the development of novel focusing elements (‘Stark mirrors’) to guide beams of Ps atoms [130]. Such long-lived Ps beams could facilitate new avenues of research including study of the interaction of positrons with earth’s gravity and new Ps precision spectroscopy measurements.

**The many-electron, many-positron system.** A cartoon of the phase diagram of this system is shown in figure 18 [128]. Most of the phases have yet to be created and studied. An exception is that, using a specially tailored pulsed positron



**Figure 18.** Schematic (only) phase diagram of the electron–positron system as a function of density  $n$  and temperature  $T$ . The density  $n_{MI}$  of the metal–insulator transition is indicated. While this quantum phase is beyond current technology,  $Ps_2$  has been created and studied, and near-term studies of a Ps BEC and a classical pair plasma are possible. For the future, accurate calculations of key features of this phase diagram would be very useful.

beam, sufficiently dense gases of positronium atoms have been created at the surface of a solid to conduct the first studies of the Ps molecule,  $Ps_2$  ( $e^+e^-e^+e^-$ ) [131]. Efforts are underway, using advanced versions of this technology, to create a Bose-condensed gas (BEC) of Ps atoms in cavities close to the surface of a solid [132]. Experiments are also in progress to create a classical pair plasma in a levitated, superconducting magnetic dipole trap [133].

**Positron-atomic physics.** Traps and high-resolution trap-based beams have elucidated new features of positron–atom and positron–molecule interactions, including state-resolved cross sections for electronic and vibrational excitation by positron impact, studies of positron attachment to molecules via excitation of vibrational Feshbach resonances, and the measurement of positron–molecule binding energies [110, 128].

Many of the successes thus far are the result of the development of new technology. The workhorse in the field is the so-called Penning–Malmberg (PM) trap, which uses a uniform, solenoidal magnetic field (strength 0.01—several tesla) to confine particles radially and an electrostatic potential to restrict their motion in the axial direction [128]. A ‘buffer-gas trap’ (i.e. multiple PM trapping stages at successively lower gas pressures) is used to accumulate positrons efficiently from a source of low-energy antiparticles. For larger particle numbers and longer storage times, the positrons are transferred into a high magnetic field and ultra-high vacuum (UHV), where they cool by the emission of cyclotron radiation. In this way, sufficient antiparticle densities can be achieved so that the charge cloud is in the plasma regime [128, 134].

Methods have been developed to compress these single-component plasmas radially using rotating electric fields. Long confinement times (e.g. days, weeks or longer) and high plasma densities are achievable using this technique. In some cases, evaporative cooling is used to lower the plasma temperature even further [135], and autoresonant drive of

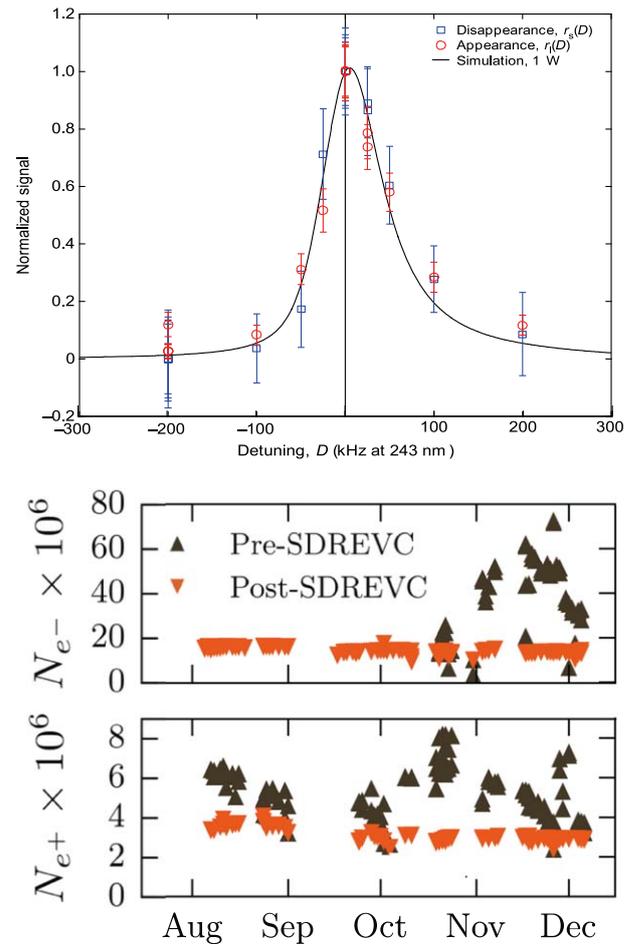
nonlinear modes of oscillation of the plasma has been used to transfer, with precision, collections of antimatter from one location to another [128]. An example of the power of such techniques is illustrated in figure 19. The 1S–2S interval in antihydrogen has now been measured with a relative precision of  $2 \times 10^{-12}$  [129]. The recent advance (lower panel), that enabled these measurements [135], was the use of rotating electric field compression and evaporative cooling (dubbed the SDREVC technique) to increase plasma reproducibility. This increased the formation rate of trappable antihydrogen by more than an order of magnitude.

*Challenges and the advances in science and technology needed to meet them.* There is good reason to believe that such tailoring of antimatter plasmas in the laboratory is in its infancy. In looking to the future, specific needs will vary with the intended goal. Following are some outstanding scientific and technical challenges that would help to achieve specific goals.

*Short temporal bursts of positrons.* Positron annihilation lifetime spectroscopy (PALS) is a workhorse in the study of materials, where lifetimes vary from a few 100 ps to nanoseconds. Ordinarily, such measurements are made in a single-count regime, with a timing start signal obtained when a fast positron is emitted from the source and the stop signal from the detected annihilation gamma ray. It would be an immense step forward if one could deliver a burst of positrons in a time of the order of 100 ps and use the burst as the start signal [128].

*Colder positrons and antiprotons.* Antihydrogen formation and trapping relies on cold antiprotons, cold positrons, and cold antihydrogen atoms. The antihydrogen production rate is temperature sensitive, and the neutral antihydrogen trap depth is very small ( $<0.6$  K). The development of improved cooling methods would facilitate significant increases in antihydrogen trapping rates.

*Higher positron densities.* As mentioned above, a major challenge on the horizon involves study of the multiple phases of the many-electron many-positron system illustrated in figure 18. The first study of this kind focused on the positronium molecule,  $\text{Ps}_2$  [131]. Work is now underway to study the much more challenging quantum many particle state, the Bose Einstein condensate (BEC) of Ps. The current approach involves directing higher areal density (i.e. positrons/per unit area) beams to a material surface, which could be accomplished using higher density plasmas in electromagnetic traps [132]. One can approach the theoretically limiting ‘Brillouin’ density  $n_B$  in positron traps at low magnetic fields using buffer gas cooling; but the low field severely limits the density. Unfortunately, the fraction of the  $n_B$  limit currently achieved at tesla-strength fields is orders of magnitude smaller [128]. Improving this apparently extrinsic impediment would be an immense help in the BEC quest and other experiments.



**Figure 19.** A success of advanced trapping techniques: (above) two-photon spectrum of the antihydrogen 1S–2S transition, measured with a relative precision of  $2 \times 10^{-12}$ ; (below) stability of the positron and electron (cooling) plasmas used to create the atoms before and after plasma tailoring by radial compression and evaporative cooling (SDREVC). The resulting stability led to more than an order of magnitude increase in the formation rate of trappable antihydrogen. Adapted from [129, 135]. Reprinted with permission from Springer Nature [129], CC-BY 4.01. Reprinted with permission from [135], copyright 2018 by the American Physical Society.

*Higher capacity traps.* In the classical regime of figure 18, the electron–positron (‘pair’) plasma will require the delivery of a very large numbers of positrons ( $10^{10}$ – $10^{12}$  or more) to the pair-plasma trap. Unfortunately, accumulation of larger number of particles creates a larger space charge and hence requires a larger confining potential. While  $10^9$  or  $10^{10}$  particles can be confined with reasonable potentials ( $\sim 1$  kV), the classical pair plasma appears to require a novel approach. One technique, currently being pursued, is to arrange multiple PM traps in parallel in the same magnetic field and vacuum (i.e. a multicell trap) [128]. An arrangement of  $p$  cells, for example, would reduce the required confinement potential by a factor of  $p$ .

*Finely focused and electrostatic beams.* Trap-based versions of such beams would enable new physics. The Ps BEC requires a high areal density. If one could convert the

magnetically guided beam from a positron trap into an electrostatic beam, other techniques (such as ‘remoderation’) could be used to increase the areal density [128]. Such an electrostatically guided beam would also enable more precise and/or higher signal-to-noise atomic-physics scattering studies.

*Advanced trap technology at intense-beam facilities.* Almost all of the work described above is done with radioisotope positron sources (commonly  $^{22}\text{Na}$ ) and solid neon moderators. Much could be gained by outfitting high-flux reactor- or LINAC-based positron beams with traps to create positron plasmas and beams tailored for specific applications.

*Portable antimatter traps.* Such traps could enable experiments with antiparticles at locations remote from high-flux sources. A trap containing  $10^{12}$  positrons would provide a continuous positron beam of  $10^6 \text{ s}^{-1}$  (similar to typical radioisotope beams) for 10 days. This would, for example, be advantageous at facilities where it is not feasible to site a dedicated positron source (e.g. at a synchrotron).

The RW technique can be used to achieve ‘infinite’ confinement times for antiprotons or positrons in a UHV trap. The practical limitation will likely be annihilation, which is

acceptably long for positrons, but will require a very good cryogenic vacuum for antiprotons [134]. Barring advances in magnet technology, the trap would also require cryogenic cooling for superconducting magnets.

*Concluding remarks.* The development of traps for large numbers of positrons and antiprotons began in the 1980s. A more or less steady stream of improved approaches, new techniques, and increased capabilities have followed. The generation of interacting Ps, the matching of laser pulses to bursts of Ps atoms, and the SDREVC technique (shown in figure 19) are arguably among the more important recent developments. The many challenges described here represent a relatively broad range of scientific and technological opportunities that can be expected to keep this area active and productive for many years to come.

## Acknowledgments

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## 14. Electron and positron interactions

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*Status.* Despite the large body of work examining positron and electron collisions there are still many outstanding problems that have not been able to be resolved. Indeed, there are still only a few systems where the cross section set for collisions from a single target could be said to be benchmarked, due fundamentally to two reasons:

1. A complete *ab initio* theory for scattering from even simple targets remains a significantly complex problem.
2. Experiments are typically unable to accurately provide cross sections for the complete set of scattering processes.

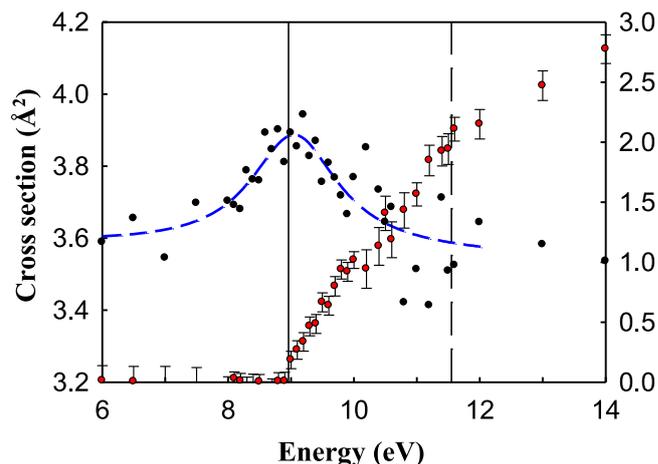
This leads to the necessity of a close relationship between experimental and theoretical efforts in order to take advantage of the relative strengths of each endeavour.

Both electron and positron scattering have made significant progress in the last 20 years with a range of new techniques able to expand the details of our knowledge for a range of kinematics and projectiles. In particular, the advent of the Surko positron trap and beam system [136] has transformed the possibilities for positron scattering measurements, leading to strong advances in the understanding of this field, as well as throwing up new challenges for calculations of positron interactions. In addition, there are a wide range of predictions from theory that remain to be experimentally verified, and will require yet further advances in technology to enable these attempts.

*Current and future challenges.* Positron scattering, in particular, remains behind electron scattering in terms of both experimental investigations and theoretical advances, but there are still significant areas in both fields for future investigations.

In the case of positron scattering, there have been numerous predictions of scattering resonances and bound states in atomic systems [137], none of which have been experimentally verified. In contrast, resonance behavior is well established in low energy scattering from molecules, and was first observed in experimental measurements [138], with a theoretical basis following on later. Most of the atoms that have been calculated to support a bound positron state are metallic, and thus provide particular difficulties for experiment, so new techniques must be developed to make these investigations. In this, positron scattering can borrow from electron scattering where investigations from metal vapours have been undertaken for many years, including a large number of observations of resonances [139].

On the other hand, there have been several cases of experimental observations that have yet to be replicated by theoretical calculations. In particular, the observation of a Wigner-like cusp at the positronium formation threshold in elastic positron scattering from noble gases [140], shown in



**Figure 20.** Observation of a threshold cusp in elastic scattering of positrons from argon (black points). Positronium formation is shown in the red points. Taken from [140].

figure 20, is not reproduced in any theoretical calculation, highlighting the difficulty of including positronium as a reaction product in the scattering calculations. Indeed, there are only a handful of calculations that include an *ab initio* description of positronium formation, although in the case of positron scattering from helium this still does not give rise to the effect observed in experiment [141]. Explicit inclusion of positronium formation remains one of the big challenges in positron scattering, from a theoretical point of view.

Even for the relatively simple case of threshold positron ionization, there is disagreement between the predictions of Klar and later work with experiment. In this case, the Wannier threshold behavior for double (positron + electron) escape from the residual nucleus is calculated to give a cross section rising as  $E_{\text{ex}}^{-n}$  near threshold, with  $n = 2.65$ , in contrast to the case for double electron escape ( $n = 1.127$ ). Indications from experimental measurements (including recent work at high energy resolution [125]) suggest that for positrons  $n \sim 1$ , in agreement with the exponent in the electron case. This remains an unresolved disagreement.

Recent experimental work on positronium scattering suggests that there is interesting and unexpected physics at play in positronium collisions from both atoms and molecules, which remains mostly unexplained by theoretical calculations (see section 12 of this roadmap for further discussion). Further refinement of both experimental approaches to positronium beams and to calculations of positronium scattering will be imperative to advance the progress of this significant new area of exploration.

Borrowing from electron scattering, the development of a positron reaction microscope would enable new insight into the details of positron scattering. As a hugely successful instrument for a range of photon, electron and ion induced ionization processes [13], positrons are the next obvious projectile that would benefit from the huge amount of detail available from this experimental technique.

In the case of both positron and electron scattering, moving away from single targets to measurements in the liquid phase is an emerging challenge for both experiment and

theory. In particular, a large amount of work in recent years highlights the importance of sub-ionisation energy electrons being responsible for a large amount of damage in DNA molecules as a result of radiation exposure [80], and moving towards a proper description of these processes in soft condensed matter will be critical to applying this knowledge in medical and radiation physics. The same can be said for positron interactions in liquid media, as work proceeds towards a better understanding of positron thermalisation processes in positron emission tomography (PET) scans.

*Advances in science and technology to meet challenges.*

There are a number of experimental obstacles to be overcome if the challenges outlined above are to be met. There is most work to be done on the positron experimental processes, due in no small part to the low flux of high energy resolution positron beams, and the limitations this puts on the measurements. An example is the development of a positron reaction microscope. While this is something that has been successfully operated with electron beams for many years [13], and with relatively low electron currents, the fact that positron sources provide beams with currents that are orders of magnitude lower limits their application to this type of experiment. Nonetheless, several attempts have been made, without measuring a triply differential cross section. It remains a challenge to develop a positron beam that can be successfully applied to these measurements and improvements in flux and intensity are likely to be needed to make this viable. Improvements along these lines will also provide the possibility for important improvements in positronium beam technology, through approaches such as the charge exchange-type approach developed by the University College London group [142]. If this can be done while retaining the energy resolution that is currently feasible using a Surko trap, further details of positronium scattering from molecules can be explored. New reactor-based positron sources may be able to provide a part of this solution, and there has been work on long-term storage of high numbers of positrons (for subsequent use in intense positron beam formation) from several groups, although a prototype that produces a suitably improved positron beam has not yet been constructed.

Other examples from electron scattering are likely to be easier to adapt, such as the introduction of metal vapour ovens. In fact, there have been previous measurements of positron scattering from targets that are solid at room temperature, and thus requiring heating to form a gas phase target, but none that have targeted the measurement of positron bound states, which require a high energy resolution, and likely a somewhat different experimental approach.

Of relevance to both positron and electron scattering is the development of liquid targets and experimental techniques that will allow for measurement of relevant parameters. In particular, being able to compare to theoretical calculations of thermalisation processes in liquid targets (and the resulting damage that takes place to DNA molecules, for instance) will enable a significant step forward in our understanding of both electron and positron scattering processes *in vivo*. This presents a non-trivial problem in many senses. Typical experiments are carried out in vacuum conditions, which mitigates against the use of a liquid as a target, especially with such a high vapour pressure as water. While some experiments have been already performed on ice targets, this is far from a perfect solution given the differences in interactions between the solid and liquid phase. There is also the question of what particular quantities can be measured usefully, to give insight into the scattering processes and to validate (or otherwise) the predictions of theory and modelling. One solution may be the extraction of projectile beams from vacuum systems, after the appropriate conditioning. This is an approach that has been successfully used in application to a pulsed positron beam [143], and provides the possibility for an ex-vacuo liquid target to be used in experiments. A similar approach would certainly be feasible for electrons.

*Concluding remarks.* While both positron and electron scattering are now mature fields of research, there are still many areas of future study to be considered. A range of new experimental approaches need to be developed to accommodate this, and theory needs to also evolve to allow for accurate solutions to complex and multi-body scattering problems. While some of these challenges are considerable, progress towards their solution will open up the study of a range of interesting and significant research problems.

## 15. Positrons for fundamental science and applications

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*Status.* Positron interaction with ordinary matter is not only an interesting and challenging subject to study. Owing to their annihilation gamma-ray signal, positrons are also unique probes of matter over a huge range of scales, from that of the whole Universe to the subatomic scales. At the upper limit, positrons are part of the matter–antimatter paradox of the visible Universe. At the lower limit, the positron is a fundamental constituent particle of quantum electrodynamics (QED) and the Standard Model. In between, positrons allow probing of matter on the scale of our galaxy, studies of the Sun, laboratory studies of materials, investigation of living bodies through positron emission tomography (PET), and probing of atomic and molecular interactions and our understanding of them, including electron correlation effects and vibrational dynamics.

### *Current and future challenges*

*Search for dark matter.* Measurements of high-energy spectra of cosmic-ray electrons and positron by satellites produced surprising results. Results from the PAMELA experiment [144] indicated an anomalous abundance of positrons with energies 1.5–100 GeV. High-energy positrons in cosmic rays originate from ‘secondary sources’ (i.e. due to interactions between cosmic-ray nuclei and atoms in the interstellar medium), as well as ‘primary sources’, such as pulsars and microquasars or dark matter annihilation. The increase of the positron fraction in the total flux of electrons and positrons above 1.5 GeV points to the contribution of a primary source. This discovery stimulated a flurry of theoretical activity regarding the question of whether such enhancement could be due to annihilation of a type of dark matters known as WIMP (weakly interacting massive particles) or other extensions of the Standard Model. Subsequent measurements with the Alpha Magnetic Spectrometer (AMS-02) at the International Space Station (ISS) extended the range of the measured cosmic-ray electron flux to 700 GeV and positron flux to 500 GeV. They found the positron fraction to be steadily increasing between from 10 to ~250 GeV. The changes in the behaviour of the electron and positron fluxes with energy, contains important information on the origins of cosmic-ray electrons and positrons. In 2017 new results became available from the Calorimetric Electron Telescope (CALET) installed on the ISS, extending the total (electron + positron) spectrum to 3 TeV, with observations of a possible structure above 100 GeV. The most recent measurements by the Dark Matter Explorer (DAMPE) measured the cosmic-ray electrons and positrons from 25 GeV to 4.6 TeV with increased energy resolution and low background [145]. These data revealed a ‘spectral break’ above 0.9 TeV, and

gave prospects of narrowing down the parameter space of models for candidates of dark matter particles that are invoked to account for the ‘positron anomaly’.

*Galactic positrons.* A most detailed experimental study of positrons in the Milky Way was performed with the gamma-ray spectrometer SPI on ESA’s INTEGRAL satellite [146]. It measured photons between 20 keV and 8 MeV using Ge detectors, with a spectral resolution of ~2.2 keV at 662 keV, for 11 years, starting in 2002. The data provided a clear picture of the dominant annihilation line at 511 keV, with a characteristic ortho-positronium (ortho-Ps) continuum on the low-energy side. It puts the total annihilation rate in the Galaxy at  $(3.5\text{--}6) \times 10^{43} \text{ e}^+ \text{ s}^{-1}$ , with 60% events in the form of a faint emission from the disk and 40% from a small bright region in the bulge. The origin of this puzzling morphology has been the subject of much speculation regarding the source (or sources) of the positrons, their transport and the properties of the interstellar medium in which annihilation occurs. Positron production is assumed to be due to radioactivity from the  $\beta^+$ -decay of  $^{26}\text{Al}$  in massive stars,  $^{44}\text{Ti}$  in core-collapse supernovae, as well as supernovae of type Ia or low-mass x-ray binaries. The numbers and distribution of such sources and their positron yields can be estimated. However, the results do not easily agree with the observations. This leaves room for other exotic positron sources, such as dark matter annihilation that would be concentrated in the central region of the Galaxy, though other explanations suggest more conventional sources [147]. The shape of the 511 keV and the adjacent ortho-Ps continuum, provide important information on the interstellar medium (ISM) where the annihilation occurs, and on the fraction of events in which positrons annihilate with atomic electrons ‘in flight’ versus annihilation through Ps formation. While most of the atomic matter in the ISM is hydrogen, specific properties of other species that might be present at small abundances can influence the outcome. For example, for hydrogen, the Ps formation channel is open only for positrons with energies greater than 6.8 eV. This means that positrons that slowed down below this energy will annihilate ‘in flight’. In contrast, for alkali atoms, such as sodium, the Ps formation channel is open at any positron energy, and its presence at solar abundance in the warm ISM can reduce the annihilation timescale for low-energy positrons by at least an order of magnitude [148].

*Positrons in solar flares.* When protons,  $\alpha$ -particles and heavier ions are accelerated in solar flares, they interact with the solar atmosphere and produce radioactive nuclei which decay by emitting positrons. Positrons are also produced in decays of positively charged pions created by protons with energies above 200 MeV. Measurements of annihilation radiation provide information on the  $3\gamma/2\gamma$  ratio, 511 keV line width and time profile of the radiation, which in turn depend on the temperature, density, and composition of the medium where the positrons slow down, form Ps and

annihilate [149]. Modeling the annihilation line parameters requires atomic data for Ps formation, as well as its break-up via charge exchange and ionization. Interestingly, the results of such modeling do not point to a single location where positron–electron annihilation takes place that would explain all the observations and be consistent with the current understanding of the solar atmosphere.

*Tests of QED.* The positron is a constituent particle of Ps, an ‘atom’ whose properties are described by QED without complications that arise for other atoms, such as the finite nuclear size. Quantities that allow direct comparison between experiment and theory are the lifetimes of ortho-Ps and para-Ps and energies, e.g. the ortho-Ps–para-Ps ground-state hyperfine splitting. On the side of theory, all energy contributions through terms of  $O(m\alpha^6)$  (in relativistic units) are known and efforts for determining next order corrections are ongoing. The precision of experimental data for both the annihilation rates and energy intervals lags behind theory. However, new precision measurements are made [150], with reduced systematic uncertainty from positronium thermalization. (Similar effects were also at the heart of the long-standing ‘orthopositronium lifetime puzzle’, with  $5\sigma$  discrepancy between theory and experiment, that was resolved in favour of theory in 2002.) Positron thermalization rates are determined by the size of its momentum transfer cross sections with the ambient gas. These cross sections are difficult to measure, even indirectly. They are also hard to predict theoretically, even for simple targets, such as noble gases, because of the need to account for the structure and dynamic polarization of the projectile and target.

*Positron emission tomography.* Conventional PET is based on coincident detection of two gamma rays from positron annihilation, with the positrons delivered by short-lived  $\beta^+$ -radioactive isotopes that tag biologically active tracer molecules. This detection establishes the line of sight and allows reconstruction of the image in the plane through computed tomography. All other information carried by the gamma rays, e.g. their Doppler shifts that can be specific to the environment in which positrons annihilate, is currently wasted by commercial scanners. However, there are prospects of new developments that may add ‘new dimensions’ to the PET images. For example, J-PET, developed by the Jagiellonian University in Kraków, uses plastic scintillators that have superior time resolution ( $\sim 100$  ps), compared to the conventional inorganic crystal detectors. This allows reconstruction of annihilation events by measuring the time-of-flight delays. It also allows reconstruction of  $3\gamma$  ortho-Ps decays and studies of the corresponding 3-photon entanglement [151]. The system also holds prospects of performing Positrons Annihilation Lifetime Spectroscopy (PALS) in living organisms, which could potentially discriminate between healthy and tumor cells.

*Positron spectroscopies in materials science.* This is probably the area where positrons make the largest impact on technology. Positron annihilation in materials and methods such as PALS and ACAR (angular correlation of the annihilation radiation) enable identification of vacancy-type and other defects in semiconductors [152] and other solids, studies of free volume in polymers and determination of sizes of nanopores, investigation of phase transitions in microvoids, reconstruction of Fermi surfaces, and surface studies, including methods such as positron-annihilation-induced Auger electron spectroscopy (PAES), which has superior signal-to-noise properties, compared with electron-induced Auger, and reflection high-energy positron diffraction (RHEPD), which has high sensitivity to the topmost atomic layer at the surface. Extracting maximum information from these techniques requires advanced theoretical methods to describe positron states in these systems, rates of positron trapping by defects and corresponding annihilation rates, etc. Currently, density-functional methods seem to dominate the field, but there is room for developing many-body theory methods, which were very important in the early years for understanding correlation effects for a positron in an electron gas.

*Positron annihilation in molecules.* This topic has seen much progress in the past 20 years, due to the development of high-quality trap-based positron beams that enabled energy-resolved measurements of annihilation rates, thereby supporting and stimulating theoretical advances. It has now been firmly established that positron annihilation in most polyatomic molecules proceeds via positron capture in vibrational Feshbach resonances (VFR). This is a reversible process in which a positive-energy positron undergoes a transition into a negative-energy bound state with the molecule, transferring its energy to one or several vibrational modes of the molecule. Positron capture increases the time it spends in an electron-rich environment, giving rise to orders-of-magnitude enhancement of the annihilation rates over those of direct, in-flight annihilation. In order to describe this process, one needs to be able to account for strong electron–positron correlations that determine the energy and wavefunction of the positron–molecule bound state. Experimentally, binding energies are now known for about 75 molecules, but only few of these have theoretical results, with best agreement currently at 25%. One also needs to understand the details of positron interaction with molecular vibrations, and the surprising efficiency with which this light projectile sets the heavy nuclear framework into motion. Anharmonic vibrational effects are key to enhanced annihilation rates, as they provide intramolecular vibrational energy redistribution (IVR), increasing the VFR lifetime against positron re-emission in some cases and mediating positron escape in others [153]. It appears that in this system, positrons act as sensitive probes of molecular vibrational dynamics. Unraveling the details of positron annihilation signal in this case has the potential of providing useful information about

IVR, a process that is of paradigmatic importance for chemical reactions.

*Advances in science and technology to meet challenges.*

Extracting maximum information in positron studies requires detailed theoretical understanding of their interactions with matter, including challenging electron–positron correlation effects in many-electron systems. In many cases, theories are only successful in simple systems. Theorists must find ways

of treating not only systems that they can treat confidently but those that are difficult and yet of great interest!

*Concluding remarks.* Being pieces of antimatter, positrons remain somewhat exotic. On the other hand, their many uses make them a household item in many areas of physics and an indispensable physicists' tool in studying the world around us.

## APPLICATIONS

### 16. Electronic collisions with atoms and molecules in astrophysics

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*Status.* Studies into the cosmic cycle of baryonic gas have long stood at the forefront of astrophysics. Much of our knowledge about this cycle rests on our understanding of the underlying electronic collisions with atoms and molecules that drive the observed properties of the Universe. However, the continual improvement in the available ground-based, stratospheric, and space-based observatories (both in telescope collecting area and in the associated spectroscopic instrumentation) means that we need to continually deepen our understanding of the relevant electronic collisions so as to keep pace with the needs of the astrophysics community in interpreting their observations.

The cosmic cycle of baryonic gas can be thought of as beginning with diffuse atomic clouds embedded in the interstellar medium (ISM) of a galaxy. As these clouds undergo gravitationally collapse, they become fully molecular diffuse clouds. These can go on to form dense molecular clouds. Some of these continue to gravitationally collapse and form prestellar cores, followed by star formation. Left over material from this process forms a protoplanetary disk surrounding the young stars. Out of this disk planets can form, some of which can have significant atmospheres. Massive stars emit copious amounts of ultraviolet radiation, photoionizing the surrounding gas to form H II regions. Stellar outflows feed material into the ISM during all phases of the life of a star. In the later stages of stellar evolution, the star photoionizes this material, forming what are known as planetary nebulae (PNe). At the end of the lifetime of a star, it forms a compact object. Stars with masses below  $\sim 10$  solar masses end their life as a white dwarf. Those more massive become supernovae, leaving behind a neutron star or black hole and forming an expanding supernova remnant (SNR) that feeds material back into the ISM. Many stars form in binary pairs. When one of the stars die, the resulting compact object can accrete material from the companion star, forming an x-ray binary (XRB). Active galaxies (AGNs) host supermassive black holes that accrete material emitting copious amounts of energy and matter back into the galaxy in the process. Stellar outflows and SNe re-initiate the cosmic cycle of gas. This cycle also feeds gas into the intergalactic medium.

Along this cycle of gas, particularly important are electron-driven processes with both neutral and charged atoms and molecules. These processes include excitation, ionization, recombination, and association. Some recent reviews of electron-driven processes in general have been given by [154, 155] and in specific for astrochemistry by [156] and for astrophysics by [157, 158]. Here we review a

few of the current and future challenges for the field. We then present some of the experimental advances planned to meet these challenges, a discussion biased by my experimental background and lack of expertise in the important theoretical work also being done in this area. This is followed by a brief conclusion. We also apologize in advance for the wide range of important work and issues that have been left out due to lack of space.

*Current and future challenges.* One of the many challenges that lie at the forefront of molecular astrophysical research is tracing the initial steps leading to star formation. This formation process can occur only after an interstellar clouds transitions from diffuse atomic gas (primarily H) to diffuse molecular gas (primarily H<sub>2</sub>). Observing this transition is challenging as H<sub>2</sub> lacks a permanent dipole moment and direct detections are extremely difficult. Also, the commonly used H<sub>2</sub>-surrogate CO is not an option as diffuse clouds provide insufficient shielding from the interstellar radiation field and most of the CO is photodissociated. In its place, observers use simple molecules that form during the transition, such as CH<sup>+</sup>, OH<sup>+</sup>, SH<sup>+</sup>, HCl<sup>+</sup>, and ArH<sup>+</sup>. But reliably interpreting the observed spectra from these molecules requires an accurate understanding of several key electron-driven processes. Electron collisions play a role in determining the populations of the various rovibrational states. The ions can also undergo dissociative recombination with the electrons, affecting the predicted molecular abundances [159].

An exciting finding over roughly the past decade has been the discovery of molecular anions in space [160]. Spectroscopic detection has been made of C<sub>2n</sub>H<sup>-</sup> anions ( $n = 1-4$ ) in the cold circumstellar envelope of the carbon-rich asymptotic giant branch (AGB) star IRC+10216 and in dense molecular clouds. Similarly, C<sub>2n-1</sub>N<sup>-</sup> anions ( $n = 1-3$ ) have also been found in IRC+10216 but not in dense clouds. *In situ* measurements by the Casini mission discovered anions in the upper atmosphere of Titan with mass-to-charge ratios from 10 to 10 000. Our understanding of the properties of these anions and the influence that they have in these molecular environments is currently limited by our lack knowledge of the fundamental processes that form and destroy these molecular anions.

Cosmic atomic plasmas can be divided into two broad classes: electron-ionized and photoionized. Electron-ionized plasmas are formed in objects such as the sun and other stars, SNRs, galaxies, and the intercluster medium in clusters of galaxies. Photoionized plasmas are formed in objects such as PNe, H II regions, XRBs, and AGNs. Understanding the spectral and thermal properties of these objects requires an accurate knowledge of the ionization level of the gas. This in turn depends on a reliable understanding of the underlying atomic ionization and recombination processes that determine the charge state distribution (CSD) [161, 162].

Another fundamental parameter controlling the behavior of cosmic atomic plasmas is the electron density. Accurate density measurements are needed to reliably determine and model the properties of these astrophysical plasmas. Spectroscopic diagnostics are used to infer the electron density from the ratio of emission lines, at least one of which is density sensitive. These diagnostics are based on theoretical calculations that can, however, differ from one another by up to an order of magnitude or more, as has been seen in recent solar coronal observation [163]

Astrophysical atomic plasmas can also be highly dynamic and are not necessarily in thermal equilibrium. Our awareness of just how dynamic and out of equilibrium these plasmas can be has only increased with every new satellite mission and corresponding improvement in the spatial and temporal resolution. However, our understanding of relevant atomic physics that controls the observed properties of object has not kept pace with these instrumental improvements. For example, for plasmas in collisional ionization equilibrium, i.e. those that can be described by a Maxwell–Boltzmann electron energy distribution, the ionization is dominated by electron impact single ionization (EISI). But for dynamic plasmas and out-of-equilibrium plasmas, electron impact multiple ionization (EIMI) becomes important. Unfortunately, the existing EIMI database is incomplete and has large uncertainties [164].

#### *Advances in science and technology to meet challenges.*

The temperature in diffuse and dense molecular clouds ranges over  $\sim 10$ – $100$  K. The recent development of the cryogenic storage ring (CSR) enables diatomic molecular ions with dipole moments to be stored long enough for them to approach thermal equilibrium with the  $\sim 10$  K chamber walls [165]. This is to be contrasted with previous generations of heavy ion storage rings where the chamber walls were  $\sim 300$  K. With the recently installed co-propagating electron beam at CSR, it will soon be possible to measure dissociative recombination on internally cold molecular ions under conditions similar to those of diffuse and dense clouds.

Interstellar anions are predicted to form via radiative association. Recent advances in laboratory techniques have enabled the study of the time-reverse process of photodetachment for some of these ions. For example, cryogenic radiofrequency tests have enabled photodetachment studies of molecular cations such as  $\text{CN}^-$  and  $\text{C}_3\text{N}^-$  [166]. Similar studies can also be carried out using CSR [167]. These methods offer the potential for improving our knowledge of the process by which interstellar anions form.

Our understanding of cosmic atomic plasmas will also be advanced by studies using CSR, which has storage times that are two orders of magnitude longer than those that were possible with the previous generation of heavy ion storage rings [75]. CSR will thereby enable near-neutral ions with long-lived metastable levels to radiatively relax to the ground state before beginning data acquisition. Using the electron beam at CSR, this will allow for unambiguous measurements of two key electron-driven processes that determine the CSD of cosmic atomic plasmas, namely electron impact ionization and the electron–ion recombination process known as dielectronic recombination.

Emission line density diagnostics can be tested experimentally using electron beam ion traps. These measurements can be used to guide theoretical calculations and to also put error bars on the accuracy of the density diagnostics. A brief review is given in [168] and references therein.

The last advance that we touch on relates to measurements of EIMI for dynamic and out-of-equilibrium atomic plasmas. Recent advances in electron gun development for ionization studies using electron–ion crossed-beams configurations now enable measurement at collision energies up to 3500 eV [169]. Measurements on this apparatus will help to reduce the uncertainties in the existing EIMI database pointed out by [164].

*Concluding remarks.* The forefront of astrophysical research is continually advancing. Our understanding of the underlying electron-driven processes with atoms and molecules that produce the collected astronomical spectra needs to correspondingly deepen in order for us to accurately interpret the observed properties of the Cosmos. Meeting these needs will require ever new advances in experimental and theoretical atomic and molecular physics. Taken together, this interaction of astrophysics with atomic and molecular physics promises many more decades of rich, exciting, and cross-disciplinary research.

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## 17. Plasma spectroscopy related to fusion energy research

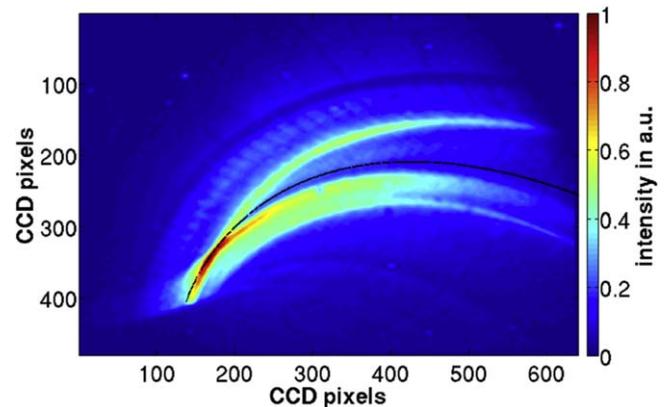
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*Status.* Accurate measurements of light emission from fusion plasmas became one of the most successful and important diagnostic techniques due to its non-intrusive nature and ability to provide temporarily and spatially resolved data. The spectroscopy of inertial confinement fusion (ICF) plasmas emphasizes x-ray emission analysis and, in particular, such effects as line broadening and opacities that are important for high electron densities,  $N_e \geq 10^{20} \text{ cm}^{-3}$ . The magnetic confinement fusion (MCF) devices, on the other hand, typically operate at much lower densities, on the order of  $10^{13}$ – $10^{14} \text{ cm}^{-3}$ , and the divertor regions in tokamaks may reach densities of few units of  $10^{15} \text{ cm}^{-3}$ . The particle temperatures vary considerably in MCF machines, from about 1 eV in the divertor to  $\sim 20$  keV in the core plasmas, and the neutral beam energies are planned for up to 1 MeV. Such diversity of plasma conditions brings about a remarkable array of spectroscopic diagnostic methods that include molecular and atomic spectroscopy in a very wide spectral range (figure 21 shows an example of a spectrum from a single line in the visible range [170]).

The amount of information that can be inferred from accurately measured spectra in MCF is voluminous and includes, for instance, spatially resolved plasma temperature and density, bulk motion, impurity concentrations and transport, strengths and other characteristics of magnetic and electric fields, to name just a few. Highly advanced, integrated spectroscopic diagnostic systems are implemented on all existing MCF machines without exceptions. The international tokamak ITER will have more than a dozen spectrometers covering near-infrared, visible, vacuum ultraviolet, and soft and hard x-ray ranges in order to fully characterize its plasma condition and evolution [171]. In the next generation tokamak DEMO, the emphasis of spectral measurements is expected to shift towards plasma condition control rather than research, yet the suite of spectroscopic instruments will be extensive.

*Current and future challenges.* A proper analysis of fusion plasma emission typically includes identification of primary spectral features and adequate modeling of the measured spectra. Such a study necessarily begins with evaluation of the relevant atomic data. While the fundamental spectroscopy of hydrogen, deuterium and, to a lesser degree, tritium as well as other relatively light elements has reached a very satisfactory level with high availability of reference data [172], this is not necessarily the case for heavier elements of importance for fusion such as Kr, Xe, and W. The spectroscopy of tungsten is of special status since this element is expected to be the major plasma-facing material (PFM) in the divertor of ITER. Although numerous studies of



**Figure 21.** C III 465 nm line emission image measured by the Doppler coherent imaging spectroscopy camera during ASDEX Upgrade tokamak shot 33 666 [170]. The black line indicates the position of the X-point. © IOP Publishing Ltd. All rights reserved.

miscellaneous spectra from various ions of tungsten brought about significant amount of data (i.e. energy levels and wavelengths), there are nonetheless ionization stages, in particular, of middle charges, for which almost no spectroscopic data exist. Besides, examination of critically important accumulation of high-Z impurities in the plasma core that will also be controlled through utilization of high-resolution x-ray imaging systems combined with XUV survey spectrometers requires precise atomic structure data.

The account of collisions in fusion devices may become rather laborious due to the variety of processes involving interactions with electrons, molecules, hydrogen isotopes, and impurity ions in the plasma. The electron collisions for the lightest elements have been a subject of extensive research, in particular, due to long-term efforts of the International Atomic Energy Agency (IAEA) and Oak Ridge National Laboratory (ORNL) to generate evaluated and recommended sets of electron-impact cross sections and/or rate coefficients [173]. Notwithstanding considerable progress, this activity is still far from completion. As for the heavier elements, the high-temperature conditions of fusion plasmas greatly alleviate calculation of basic electron interactions with highly charged ions due to reduced contribution of resonance processes. Moreover, a number of freely available atomic codes provide reasonably accurate methods for fast calculation of the relevant collisional characteristics. Yet some processes, of which dielectronic recombination (DR) for heavy elements is probably the most critical one, require additional theoretical and experimental analysis in spite of the recent advances (e.g. [174]).

In addition to electron-induced processes, interactions between heavy particles are also essential for MCF research. These collisions represent the major part of the atomic and molecular database used in plasma edge kinetic codes (e.g. B2-EIRENE [175]). The active neutral beam spectroscopy is another representative example of extremely rich diagnostics that can offer local data, e.g. on impurities and plasma fields. Here it is the collisions between the beam atoms and plasma protons and/or impurity ions that are the primary atomic

processes controlling plasma emission. The charge-exchange recombination spectroscopy (CXRS) as well as the passive spectroscopy systems in the XUV energy range require extensive and detailed information on the cross sections which are still largely unavailable for  $H(D) + Z^{q+}$  (here  $Z = Ti, Fe, Mo, W$ ).

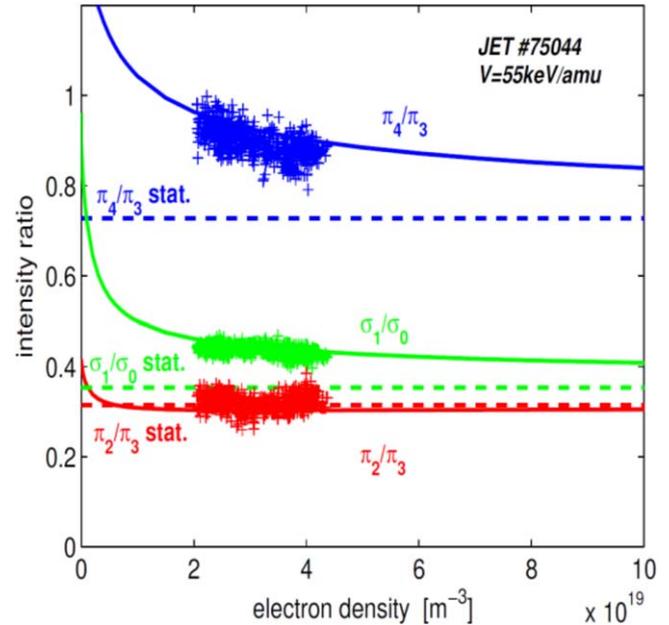
The motional Stark effect (MSE) diagnostics is based on exploration of spectra from very fast hydrogen or deuterium atoms due to their interactions with plasma particles (primarily protons) under the induced electric field of magnetically confined plasma. While the splitting of Stark multiplets can be calculated with high precision, the relevant data for collisional interactions are almost non-existent. The primary issue here is the necessity of utilization of parabolic states for the beam atoms which requires new techniques involving both cross sections and scattering amplitudes (density matrix elements). The recent comparisons with the JET data [176] and data from other tokamaks unambiguously point out to supremacy of such new methods as compared to the classical statistical models (figure 22).

The modern collisional-radiative models and codes such as ADAS [177] that make use of large-scale databases of atomic (and molecular) data are well capable of successfully simulating most of the spectral emission from MCF plasmas. Yet the ionization balance of heavy impurities in hot fusion plasmas still evades a comprehensive interpretation, likely due to computational difficulties in calculation of dielectronic recombination.

#### *Advances in science and technology to meet challenges.*

There are several directions of research where new advancements can greatly contribute to better spectroscopic diagnostics of fusion plasmas. To this end, small devices capable of producing highly-charged ions, such as electron beam ion traps (EBITs), will continue to generate a wealth of spectroscopic and even collisional, including DR, data for impurity elements and may also potentially serve as calibration sources for cutting-edge x-ray spectrometers. This effort will be complemented by measurements on the existing and new storage rings.

On the theory side, accurate calculations of heavy-particle collisional characteristics including charge-exchange cross sections and ion-neutral excitation scattering amplitudes (e.g. [178]) are expected to stimulate development of accurate CR models and significantly improve both CXRS and MSE diagnostics. As for the general collisional-radiative modeling of fusion plasmas, new DR calculations should be instrumental in improvement of the agreement between theory and measurements. It is also very important to continue intelligent



**Figure 22.** Comparison of the measured (symbols, JET tokamak) and calculated (lines) component ratios under motional Stark effect conditions [176]. Solid lines represent the results from the parabolic collisional-radiative model while horizontal dashed lines are the statistical model predictions. © IOP Publishing Ltd. All rights reserved.

comparisons of different CR codes through theoretically developed test cases as they provide a very powerful benchmarking tool. Finally, as it has been the case for many decades, generation of thoroughly evaluated and recommended sets of atomic and molecular data will remain an invaluable effort.

**Concluding remarks.** Spectroscopic diagnostics of fusion plasmas is and will be in much demand due to its unique features and advantages. As such, both theorists and experimentalists will undoubtedly continue to improve diverse diagnostic techniques through more accurate atomic and molecular data, better description of elementary physical processes, and more broad representation of plasma effects through spectroscopic imprints.

#### **Acknowledgments**

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## 18. Laser produced plasma light sources for extreme ultraviolet lithography

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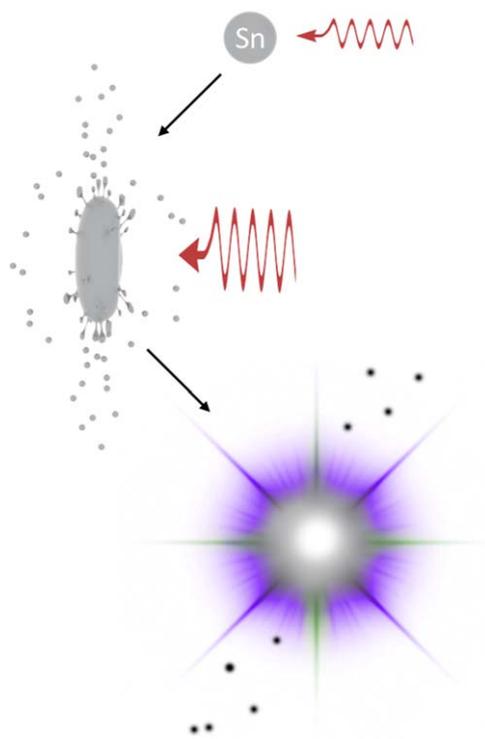
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**Status.** The use of laser produced plasmas (LPPs) as short-wavelength sources in the extreme ultraviolet (EUV) and soft x-ray spectral regions has been motivated by the application of such sources in a number of high profile areas of science and technology by far the most important of which is extreme ultraviolet lithography (EUVL). It is extreme ultraviolet (EUV) light at 13.5 nm wavelength that drives the next generation of nano-lithography machines. The step towards EUV is crucial to the continuation of the miniaturization of the features on chips. Ever since the seventies, the miniaturization is well-represented by Moore's law, which predicts that the number of transistors on an affordable CPU doubles every two years. Present-day technology uses 193 nm light. The shorter the wavelength, the better the resolution, therefore much smaller features can be printed with EUV-based lithography. Sustaining this 'law' is essential for the continued growth of the global semiconductor industry. At the time of writing, EUVL sources, operating at a wavelength of  $\lambda = 13.5$  nm are being introduced. The choice of wavelength was determined by the availability of Mo/Si multilayer mirrors (MLM) with a record reflectivity of  $\sim 70\%$  in a narrow bandwidth around 13.5 nm [179].

For a LPP generating EUV light emission from moderate to highly charged plasma ions must be involved and the source should possess a plasma temperature sufficient to produce large populations of the relevant species while the conversion efficiency (CE) of laser energy into in-band EUV at 13.5 nm should be as high as possible [180]. The three candidate elements most widely studied were Li, Sn and Xe, all of which have ions with strong resonance transitions within the required 2% bandwidth at 13.5 nm. In Li the 1s–2p doublet of H-like  $\text{Li}^{2+}$  lies at 13.5 nm but the intensity available from either laser-produced plasmas (LPPs) or discharge-produced plasmas (DPPs) was found to be too low for a viable source [181]. Although Sn plasmas were known to emit strongly at 13.5 nm, early work had concentrated on Xe plasmas since, being an inert gas, Xe posed fewer debris problems than Sn. The transitions responsible for the 13.5 nm EUV light are  $4d^8-4d^75p$  resonance lines in  $\text{Xe}^{10+}$  [182].

However, the conversion efficiency of Xe plasma sources remained lower than the ones of comparable Sn plasma based sources [180]. Near 13.5 nm, the EUV spectrum of Sn is dominated by intense transition arrays, arising mainly from resonance  $4p^64d^n - 4p^54d^{n+1} + 4d^{n-1}4f$  transitions in  $\text{Sn}^{9+} - \text{Sn}^{13+}$  [183]. Since several ion stages of Sn contribute and the line strengths for 4d–4f transitions are greater than the 4d–5p ones in Xe the maximum of the CE for



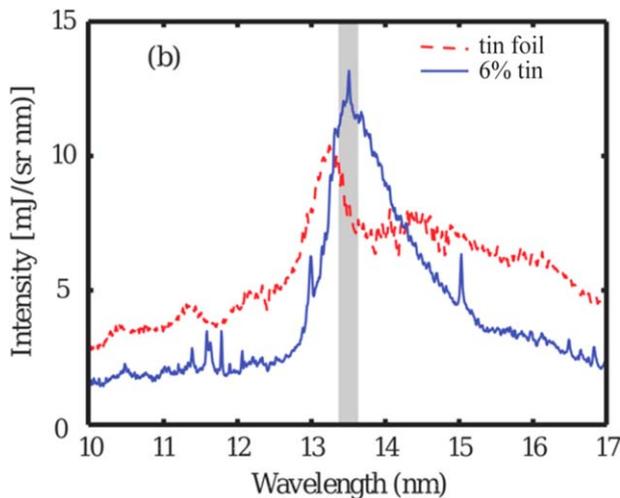
**Figure 23.** Schematics of efficient EUV generation in laser-produced plasma (LPP) sources for nanolithography using mass-limited liquid tin droplets as targets which are irradiated by infrared laser light. A laser pre-pulse reshapes the droplet into a pancake shaped target, which is hit subsequently by the main laser pulse driving the EUV generating LPP.

a Sn LPP is superior. Highly charged tin ions are thus the preferred atomic sources of 13.5 nm light.

The use of a mass-limited target, which contains the minimum number of Sn atoms needed for high-efficiency EUV emission, is required to reduce tin-debris from coating and damaging plasma facing materials. Therefore, LPP sources based on a stream of liquid tin droplets, operating at high rep rates up to  $\sim 100$  kHz, are the EUV sources of choice (see figure 23).

In an LPP most of the laser radiation is absorbed at a density below but close [184] to the critical value  $n_{ec} \sim 10^{21} / \lambda_L^2 \text{ cm}^{-3}$ , where  $\lambda_L$  is the laser wavelength in  $\mu\text{m}$ . Therefore even when using infrared lasers to drive the LPP the densities are very high and the plasmas are optically thick. Since typical LPP plasma temperatures are tens of eV the LPP plasmas are situated in a barely investigated region of the density-temperature landscape of plasmas lying in between fusion, solar, and lightning plasma.

**Current and future challenges.** The understanding, control, and optimization of LPP EUV and soft-x-ray sources in general and 13.5 nm Sn LPPs for nanolithography in particular is challenged by modelling capacities requiring high-quality data on atomic structure, atomic interactions and radiation transfer in dense plasma. The opacity issue (see figure 24) can be and large be circumvented by the use of  $\text{CO}_2$  lasers operating at  $10.6 \mu\text{m}$  instead of more compact,



**Figure 24.** Illustration of opacity affecting the EUV spectra of a Sn plasma. Blue curve: Optically thin spectra of tin doped (6%) glass. Red curve: Optically thick spectrum of a tin foil [186]. © IOP Publishing Ltd. All rights reserved.

user friendly Nd:YAG ( $\lambda_L = 1.06 \mu\text{m}$ ) since increasing the wavelength by a factor 10 reduces the critical electron density by a factor of 100. Moreover, the optical depth increases with plasma length scales, e.g. laser pulse duration or droplet size. Detailed calculations showed that using  $\text{CO}_2$  laser light incident on Sn, a CE of up to 6.5% was attainable in a 30 eV plasma [185].

Direct illumination of droplets with diameters of  $100 \mu\text{m}$  or less results in low CEs as the laser beam diameter generally exceeds that of the droplets. To overcome this problem, nowadays dual pulse irradiation is used. A low-power laser pulse (prepulse) is focussed onto the liquid tin droplet and by the generation of a pre-plasma in front of the droplet it kickstarts a fluid dynamical deformation of the droplet, reshaping the target into a thin disk ‘pancake’. The pancake-shaped target is irradiated by the main pulse laser creating the EUV generating LLP. Critical factors are inter-pulse time delay, pulse shapes, target shapes and irradiation conditions, which continue to be active topics of research in order to fully maximise conversion efficiencies.

Predictive plasma modelling requires accurate atomic data for the Sn ion stages involved. The early analysis of Sn spectra was based on data obtained from spark discharges which contained lines from many ion stages overlapped within a narrow wavelength range. Recent experimental work on ion stage differentiated EBIT (electron beam ion trap) spectra has shown that the origin of some lines was incorrect and that the spectra need to be revised [187]. High-resolution

spectral data are required to supplement the EBIT spectra for accurate determination of Slater Condon parameters for spectral fitting and to drive theoretical developments to handle complex, many-electron, open-shell systems. In addition to spectral data which are starting to become available, accurate values for excitation, recombination and collisional rate coefficients are required. Except for data on electron impact ionization [188], (experimental) data on other atomic interactions is barely present.

Current 13.5 nm EUV sources rely on  $\text{CO}_2$ -gas laser technology. This technology has significant drawbacks compared to modern solid-state lasers, e.g. user friendliness, wall plug efficiency, and spatial footprint. However, it would require significant studies into the plasma physics driven by the shorter-wavelength (IR to mid-IR) wavelengths available to make such sources industrially viable.

For the far future one has philosophized shorter wavelengths for beyond EUV lithography or soft x ray imaging applications. In particular, 6.7 nm is mentioned based on the potential availability of MLMs with a high (>80%) theoretical reflectivity. Again, the strongest emission available at this wavelength comes from open-shell 4d–4f transitions, the strongest of which are emitted in  $\text{Gd}^{17+} - \text{Gd}^{19+}$ . Since higher ion stages are involved than in Sn, a higher plasma temperature close to 100 eV is required, so a higher laser power density is required and the fraction of laser energy required for ionization is greater than in Sn. Indeed the maximum CE attainable from a Gd plasma under ideal conditions appears to be of the order of 1% [189]. The greater power densities implying tight focusing of the laser beam means that  $\text{CO}_2$  lasers are generally unsuitable. Again a move to shorter wavelength solid-state lasers (e.g. Nd:YAG) appears inevitable which makes understanding strong opacity effects in LPP plasma of great eminence.

*Concluding remarks.* The advances required can be separated in scientific and technological challenges. On the technical side, some of the specs of  $\text{CO}_2$  lasers are not on par with solid state lasers. The development of high efficiency solid state laser systems in the 1–10  $\mu\text{m}$  range would greatly help in the optimization of 13.5 nm source. For LPP sources driven by such lasers the plasma is optically thick and detailed modeling efforts are computationally very demanding and heavily rely on the availability of accurate atomic data. The production of such fundamental data is of great scientific merit as it requires present day AMO research to push forward into the regime of truly many-body quantum systems and their interactions.

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