

# Benchmark measurements and theory for electron(positron)–molecule(atom) scattering

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## Abstract

The level of accord between experiments, and experiments and theory, for a number of ‘benchmark’ atomic and molecular scattering processes is reviewed. Both electron and positron impact is considered and processes such as elastic scattering, vibrational and electronic excitation and positronium formation are reviewed.

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## 1. Introduction

The field of electron(positron) collision physics is driven by a need to uncover, and understand, the broad range of dynamical processes that can occur when an electron or positron interacts with a complex atomic or molecular species. Many of the processes that have been uncovered to date are now known to have a profound effect on the way in which many everyday technological devices operate, and many natural phenomena proceed. A range of beautiful examples, which come from electron scattering, are the resonant interactions that lead to the formation of temporary negative ions, a process which occurs in *every* atom or molecule. In molecules these transient species, which can have lifetimes as short as a femtosecond, are particularly important as they strongly mediate excitation processes such as vibrational excitation. They also lead, in many molecules, to dissociative attachment, whereby the negatively charged complex breaks up into a neutral and negatively charged fragment. In many cases the product fragments

are either stable negative ions and/or highly reactive free radicals, so these transient scattering events can often lead to important chemical processes.

While the elucidation of the dynamical aspects of these reactions is important, it is also important to understand the magnitude or rate with which they occur, in order to be able to quantify any possible applications. Thus, measurements of *absolute* scattering cross-sections and reaction rates are of critical importance across a range of scattering problems. In order to establish standards for such measurements, it is equally important that there be a series of ‘benchmark’ measurements and calculations in the field, both as a standard for experimental and theoretical practice and as a guide for how theory may be applied to the many cases where experiments are intractable, for one reason or another.

In this paper, we shall very briefly review some cases of low energy electron and positron scattering where experiment and theory are in good accord. In the case of electron scattering there are several examples where agreement between experiment and theory is generally better than 10%, and many others where it is within 20%. This applies for both differential (in angle) and integral cross-section measurements. For positron scattering the situation is not

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so clear. Advances in positron scattering have been, until recent times, largely hampered by available technology, which has limited resolution and sensitivity and resulted in relatively few absolute scattering measurements. In recent years there have been significant advances using both established experimental techniques and new ones based around trap-based beams. The added complexity of the inclusion of positronium formation in *ab initio* theoretical calculations means that there are substantially fewer scattering systems that may be considered benchmarks.

A number of extensive review articles on resonant processes [1] and electron [2,3] and positron [4] scattering are available in the literature for the interested reader.

## 2. Electron–atom collisions

In many scattering channels (elastic, vibration, electronic excitation) absolute cross-sections for electron scattering are derived with respect to the value of the elastic scattering cross-section at that incident energy and angle. In most cases, the elastic scattering cross-sections for the target atom or molecule have generally been measured using the relative flow technique (see [5] for a detailed discussion and further references), where the scattering intensity for the gas of interest is measured relative to that for a standard system, usually helium. As a result, there are many cross-sections in the literature which have the elastic cross-section for helium as their basis, and it is a prudent starting point, perhaps, to discuss how well this cross-section is known.

In Fig. 1(a) and (b) we show a couple of examples of experiment and theory for the differential elastic electron–helium cross-section at energies below the first excitation potential of the atom. In this regime, experimental angular distributions can be placed on an absolute scale using reliable phase shift analysis techniques (e.g. [5] and references therein) and they can be compared with the best available

scattering theory, including for example the ground-breaking variational calculations of Nesbet [6] and the convergent close coupling calculation (CCC) of Bray and collaborators [7] and the *R*-matrix plus pseudostates calculation (RMPS) of Bartschat [8]. In Fig. 1(a) we show a comparison between experiment [5] and theory [6,7] at an energy of 1.5 eV. The level of agreement is clearly excellent, within a few percent across the entire range of scattering angles. At 18 eV (Fig. 1(b)) there are some differences ( $\sim 5\%$ ) between experiment and theory at middle scattering angles, although the three calculations [6–8] are in excellent agreement at all scattering angles – they are largely indistinguishable in the figure. The CCC calculation that is shown is one in which the normal frozen(single)-core approach has been replaced with a 4-core calculation which gives much a better representation of the dipole polarizability, and better agreement with experiment, and the other theories at small angles. The RMPS calculation includes 5 physical and 36 pseudostates. It would seem that any of these calculations could readily be used to generate the ‘standard’ cross-sections used in relative-flow experiments. It is also worth noting another set of standard helium cross-sections, tabulated by Boesten and Tanaka [9], that is based on a hybrid of experimental and theoretical results.

For other atomic species, such as the heavier rare gases, there is also good agreement for low energy elastic scattering between experiment and a range of different theories. This is particularly the case for neon, and to a lesser extent argon and xenon, although space does not permit a detailed comparison here.

Inelastic scattering presents particular challenges when it comes to measuring absolute cross-sections. As most of these measurements are made relative to the elastic scattering intensity, a major issue to be considered is the relative response of the spectrometer for scattered electrons of greatly differing energies. At high energies this is not such

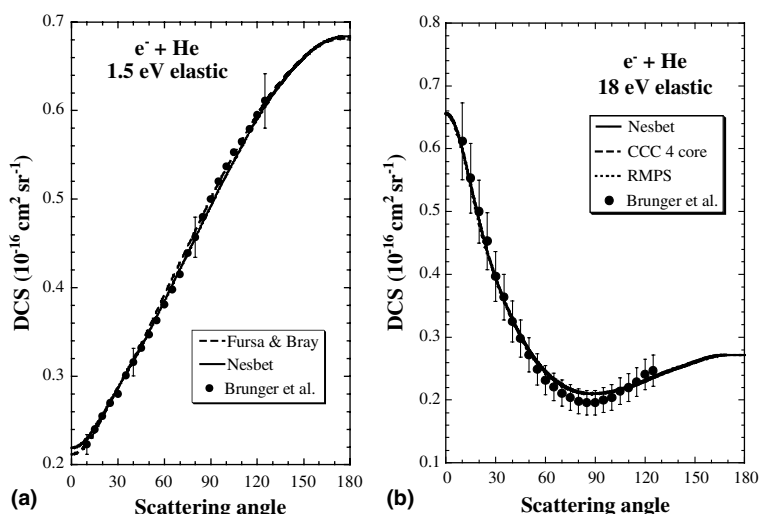


Fig. 1. Elastic DCS for electron helium scattering at (a) 1.5 and (b) 18 eV. Experimental results [5] are compared with a variational calculation [6], a convergent close coupling calculation [7] and the *R*-matrix with pseudostates calculation [8].

a significant problem as the incident and scattered electron energies are reasonably close. However, for near-threshold excitation (i.e. within a few electron volts of threshold), the elastically scattered electrons can have energies 10–100 times greater than those resulting from inelastic scattering and the response of any energy analyser is likely to be vastly different across such a range of energies. Various strategies have been adopted to overcome this issue in order to use the (usually) highly reliable elastic scattering measurements as a standard. Space does not permit a detailed discussion here but a number of groups have developed techniques to tackle these problems and excellent examples can be found in the work of Allan [10] and Khakoo [11].

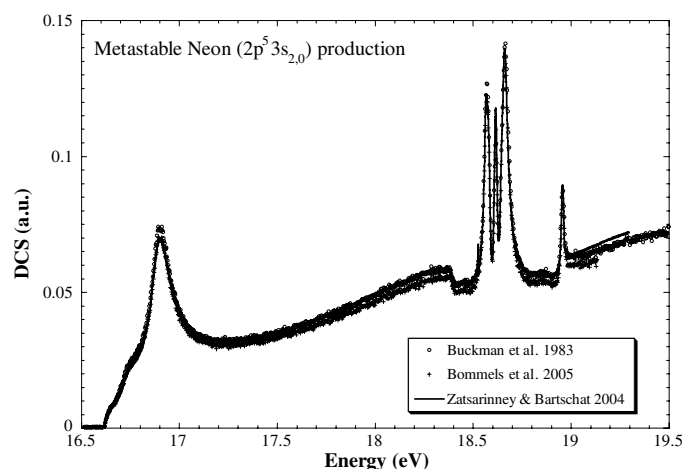


Fig. 2. Metastable atom excitation function for neon. The experimental points of [12,13] are compared with the  $R$ -matrix calculation of [12,14].

Some interesting examples of the status of near-threshold electronic excitation, both experiment and theory, can be drawn from some very recent [12] and not so recent [13] measurements on the  $2p^5 3s$  manifold of excited states in neon. In Fig. 2 we show two measurements of the metastable atom excitation function for neon within a few eV of the thresholds of the two excited states concerned ( $2p^5 3s$  ( $J=2,0$ )). These measurements are not absolute but have been normalised to each other and to the theoretical calculation which is a B-spline  $R$ -matrix approach [14]. The agreement between experiments and experiment and theory is spectacularly good, both in terms of the energy dependence of the cross-section, and the description of the negative ion resonance features which dominate it. An equally impressive comparison for differential electron excitation functions for all four members of the  $2p^5 3s$  manifold is shown in Fig. 3. The experimental measurements [15] which have been made after careful attention to details of the scattering geometry and detection efficiency, show a remarkable level of agreement with the  $R$ -matrix calculations.

Another technique which is presently being developed for benchmark near-threshold excitation measurements is a time-of-flight approach, where all scattered electrons (elastic and inelastic) are allowed to drift through a long field-free region and are detected on a large area position sensitive detector [16]. The incident electron beam is pulsed, and the time of arrival of the scattered electrons is used to determine their energy. The large area detector enables simultaneous measurements over an angular range of about  $22^\circ$ , with an angular resolution which is better than  $1^\circ$ . This technique has been used, to date, for benchmark measurements on the  $n=2$  manifold in helium which

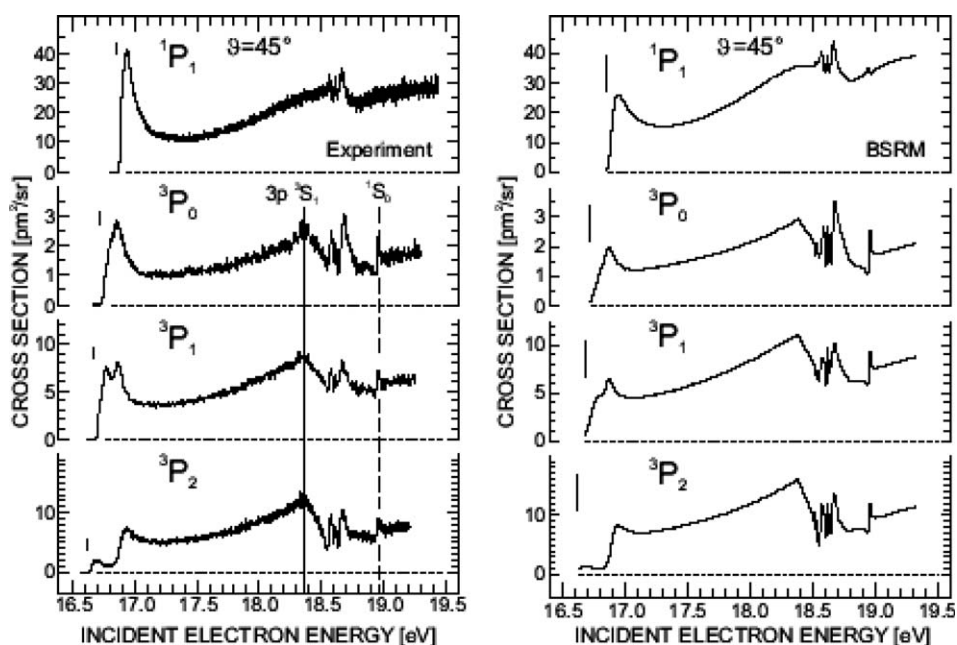


Fig. 3. Excitation functions for the  $2p^5 3s$  levels of neon at a scattering angle of  $45^\circ$ . The experimental measurements of [15] are compared with the  $R$ -matrix calculation of [14,15].

enable a close comparison with state-of-the-art CCC and  $R$ -matrix calculations.

### 3. Electron–molecule collisions

Electron–molecule scattering experiments can, in principle, be performed with the same level of accuracy as those with atomic targets. In practice however, the increased complexity of rotational and vibrational motion generally requires higher energy resolution and the reactive nature of many molecules can pose problems for stability and gas-handling. These issues are relatively minor though, when compared to those faced by scattering theory in developing state-of-the-art approaches to electron–molecule scattering calculations, particularly for polyatomic systems.

Nonetheless there have been substantial advances in the field in the past 10 years. For diatomic molecules the prototypical system for study has been  $N_2$ , both as a result of its pervasive occurrence and the low energy ( $2\text{--}5\text{ eV}$ )  $^2\Pi$  shape resonance which dominates both elastic scattering and vibrational excitation. There have been many studies of this molecule over a long period of time (e.g. [17] and references therein) and an example of the level of agreement between experiment and theory is shown in Fig. 4(a) and (b). In Fig. 4(a) we show the elastic DCS measured at an energy of  $2.61\text{ eV}$ , an energy which corresponds to the position of a quasi-vibrational resonance peak in the  $0\text{--}1$  vibrational excitation channel. In this case the original experimental values of [17] have been supplemented by data taken on the same apparatus but using a magnetic angle changer [18] in order to access large scattering angles. The agreement between the experimental measurements and the vibrational close coupling calculation of Morrison and colleagues is exceptionally good. Similarly, for the  $0\text{--}1$

vibrational excitation channel (Fig. 4(b)), we see extremely good agreement between experiment and the same theory.

For heavier polyatomic systems the situation is not so favourable when we compare experiment with theory. A case in point is the linear triatomic molecule  $N_2O$  where, at a low energy corresponding to the position of the  $^2\Pi$  shape resonance ( $2.3\text{ eV}$ ), experiment [19] and theory [20] are in poor agreement (Fig. 5(a)) for the elastic DCS. In the same molecule at higher energies ( $>10\text{ eV}$ ) we see considerably better agreement (see [19]). This molecule also exhibits an interesting feature in the angular DCS that has been observed in many other non-polar systems (see [19,21] for discussion). At energies just above that of the characteristic low energy shape resonance, many diatomic and polyatomic molecules exhibit a sharp, forward-angle minimum in the DCS, which develops and then disappears within a few eV, typically between  $3$  and  $8\text{ eV}$ . An excellent example of this occurs in  $N_2O$  and this is shown in Fig. 5(b), at an energy of  $6\text{ eV}$ . As can be seen, theory does not predict this feature, a situation which is common in almost all of the cases where it has been seen experimentally.

### 4. Positron scattering

Benchmarks in positron scattering processes are somewhat more rare than for electrons, due mainly to the inferior energy resolution and lower intensity of traditional positron sources compared to conventional electron sources. With the advent of new buffer gas traps and high resolution beams, together with new techniques for scattering in high magnetic fields, improved measurements of many processes are becoming possible [22,23]. Despite the difficulty inherent in traditional positron experiments, there have still been some studies where experiment and theory have achieved what may be described as a benchmark.

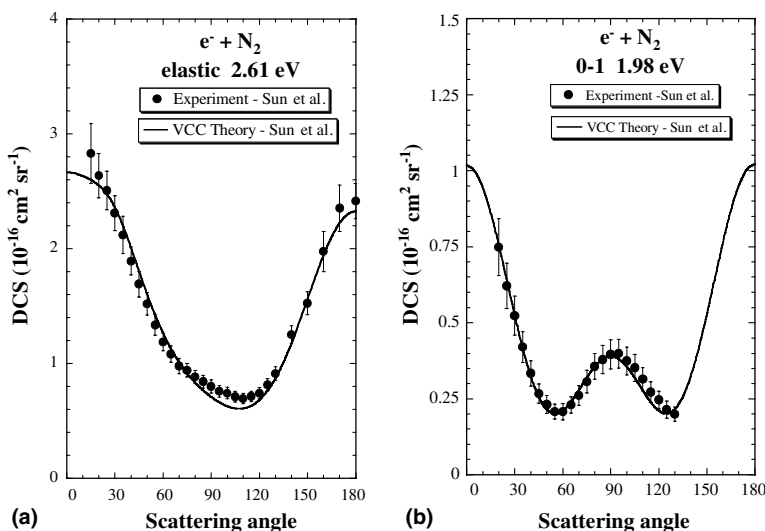


Fig. 4. Differential cross-section for (a) elastic scattering from  $N_2$  at  $2.61\text{ eV}$  and (b) vibrational excitation ( $0\text{--}1$  mode) at  $1.98\text{ eV}$ . The results are from [17,18].

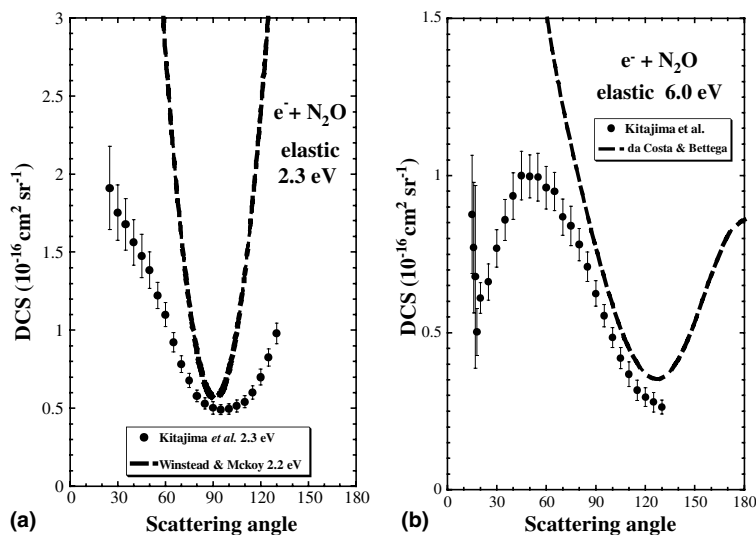


Fig. 5. Elastic differential scattering for  $\text{N}_2\text{O}$  at (a) 2.3 eV and (b) 6 eV. Experimental results [19] are compared with two theoretical calculations [20] using the Schwinger variational approach.

True benchmarks are probably limited to grand total cross-sections, where the relatively poor energy resolution of moderated positron beam sources is not a significant factor, at least for energies above a few electron volts. In particular, there are two cross-sections where the convergence of theory and experiment demonstrate the qualities required of a benchmark. These are the positron–(atomic)hydrogen and positron–helium grand total cross-sections, shown in Figs. 6 and 7, respectively. In the case of positron–hydrogen scattering, the measurements of Zhou et al. [24] are compared to various theories [25–28]. Above 6 eV incident energy, there is almost perfect agreement between all theories and the experiment. The corresponding electron scattering cross-section is also shown, and the merging of the two cross-sections can be seen at energies above about 100 eV. In the case of helium,

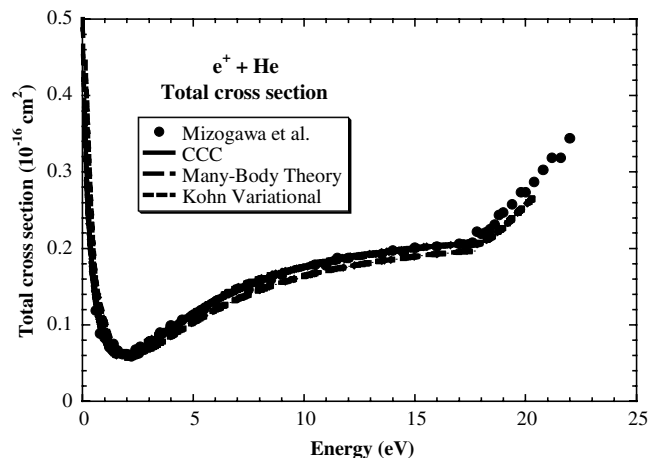


Fig. 7. Positron helium total scattering cross-sections. Solid circles are the experiments of [29], compared to a many body theory calculation [30], a CCC calculation [31] and a Kohn variational calculation [32].

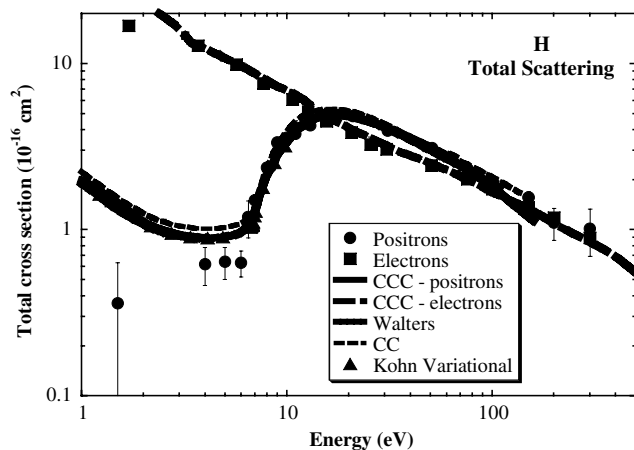


Fig. 6. Total scattering cross-sections for positrons and electrons scattering from atomic hydrogen. Circles and squares are the data of [24] for positrons and electrons respectively. This is compared to the coupled channel calculation of Walters [25], a CCC calculation [26], a close coupling calculation [27] and a Kohn variational calculation [28].

the measurements of Mizogawa et al. [29] are compared with a many-body theory calculation [30], a CCC calculation [31] and a Kohn variational calculation [32]. Below the positronium formation threshold at 17.8 eV, the agreement between theory and experiment is again excellent. Agreement between the various experimental measurements is also very good for this case, once corrections for forward angle scattering have been made [29]. This measurement also serves to illustrate a major difficulty in establishing benchmarks for positron scattering, namely positronium formation. Treatment of positronium formation is a difficult theoretical problem, yet it must be accounted for in any scattering calculation above the positronium formation threshold. This appears to have been addressed in the case of positron–H scattering, but remains to be satisfactorily solved in other systems. If a benchmark is to be defined as agreement between experiment and

theory to a certain (5–10%) level, then until the problem of positronium formation is successfully addressed in theoretical models, there will be few benchmarks for positron scattering above the positronium formation thresholds in any given system.

The issue of positronium formation is particularly relevant to the alkali metals, where the positronium threshold channel is ‘open’ at an incident positron energy of 0 eV. This means that to establish a benchmark cross-section at any energy, it must be properly accounted for. An example of a set of cross-sections for potassium is shown in Fig. 8 [33,34]. It can be seen that in this case, the elastic cross-section forms only a very small part of the grand total cross-section, except at the lowest energies, emphasising the need for the development of current theoretical models. The current theory overestimates the measured cross-section by about 10%, although the shape is in good agreement with the experimental data.

There have been very few measurements of inelastic processes in positron scattering, although recent experimental efforts are starting to open up this area of study. In particular, there are recent measurements for ionisation by positron impact in the noble gases from two different experimental groups [35,36]. In the case of positron scattering, it is convenient to define three types of ionisation; positronium formation, where a target electron is bound to the incident positron and travels away from the residual ion core; direct ionisation, which is analogous to ionisation by electron impact with both the incident and ejected particles travelling from the ion core as free particles; and total ionisation, which is the sum of the two processes. Measurements by the San Diego group measure the direct ionisation and positronium formation cross-sections, obtaining the total ionisation cross-section by adding the two [36]. In the case of the UCL group, the total and direct ionisa-

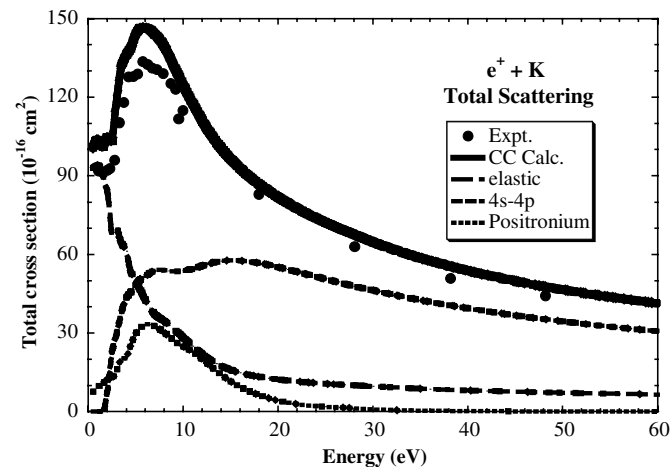


Fig. 8. Positron scattering from potassium. The circles are the total scattering cross-section measurements of [33] compared to the theory of [34] (solid line). Other symbols represent the theoretical partial cross-sections for elastic scattering (long dash), positronium formation (dots) and 4s–4p excitation (short dash).

tion cross-sections are measured, with the positronium formation cross-section the difference of the two measurements [35]. Fig. 9(a)–(c) shows the comparison of the three cross-sections in argon from the two experimental groups. The total ionisation cross-sections are in good agreement, as are the direct and positronium formation cross-sections near threshold. The direct ionisation cross-sections of the two groups are in disagreement in magnitude, although similar in shape, and this is also reflected in the significant disagreement in the positronium formation cross-section measurements. Comparisons of cross-section for the other noble gases show various levels of agreement, with the best being for positron ionisation of xenon. Calculations of

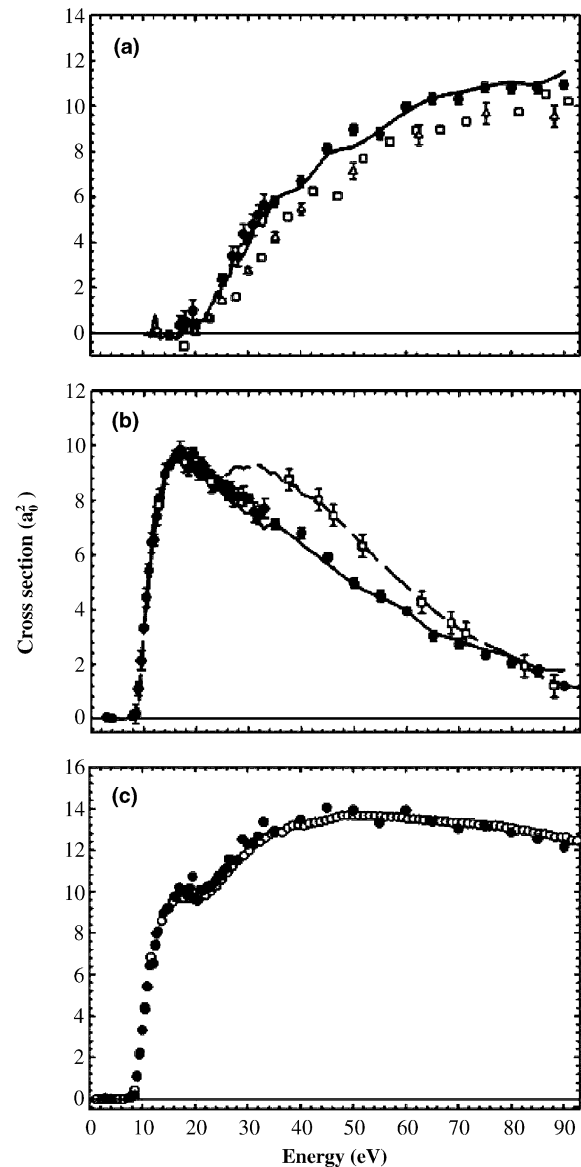


Fig. 9. (a) Direct ionisation of argon by positron impact, open symbols from [35], solid points from [36]. Solid line is from a combination of data sets from the two different groups, see [36] for details. (b) Positronium formation cross-section in argon. Symbols are as for (a), but dashed line is also from [35]. (c) Total ionisation cross-sections for argon by positron impact, symbols as for (a).

direct ionisation have been made by Campeanu et al. [37] and Bartschat [38] (argon) and show a reasonable level of agreement with the experimental measurements. For positronium formation, the agreement with available theory is worse, as may be expected. Nonetheless, for the calculations of McAlinden and Walters [39], again in argon, the magnitude of the cross-section is similar to the experimental values, although the shape is somewhat different, whereas for the calculation of Gilmore et al. [40] the shape of the cross-section is somewhat better, although the magnitude is quite different from experiment.

In terms of ionisation processes, it may be concluded that while benchmarks have not been established, there has been considerable progress towards this in the noble gases. More theoretical effort is needed and the resolution of experimental discrepancies required, in particular for the direct ionisation cross-sections. Further studies on the threshold behaviour of these cross-sections will also prove interesting.

In the case of differential cross-sections (DCS), there have been many measurements in different systems, mainly at energies above a few eV. Argon has been the most studied target and there have been measurements of many other systems, both atomic and molecular. Most of the measurements have been normalised either to theory or to total cross-sections. The exception to this has been the measurements made by the San Diego group, which are absolute, without reference to any other cross-section [41]. In this case, however, the DCS is ‘folded’ around 90°, which causes a loss of information, although a comparison can be made with other results that have been similarly folded. In most of the work on the noble gases, there is at least reasonable agreement in the shape of the DCS at low energy between experiment and theory. The absolute measurements of the San Diego group show good agreement with theory between 1 eV and 2 eV for positron scattering from argon and krypton [41–44]. For molecules there are few measurements, and only two published absolute cross-sections, for H<sub>2</sub> at 0.5 eV [45] and CO at 6.75 eV [23]. Relative cross-sections for a variety of molecules, at selected energies have been made by the Detroit group [46,47]. There is very little theory with which to compare the experimental measurements, and it cannot be said that benchmarks have been established for any molecular system.

Measurements of discrete excitation cross-sections are rare in positron scattering, due mainly to limitations in energy resolution. Recently there have been various measurements for total vibrational and electronic excitation [48,49], which can be compared to the few theoretical calculations that have been published for these processes. The case of vibrational excitation of H<sub>2</sub> serves to illustrate the current status of this type of measurement. The comparison between experiment and the latest theory (Fig. 10) is very good [48,50–52], although a more detailed investigation of the threshold region is warranted. Both further experimental measurements and theoretical calculations are needed before the strict definition of a benchmark

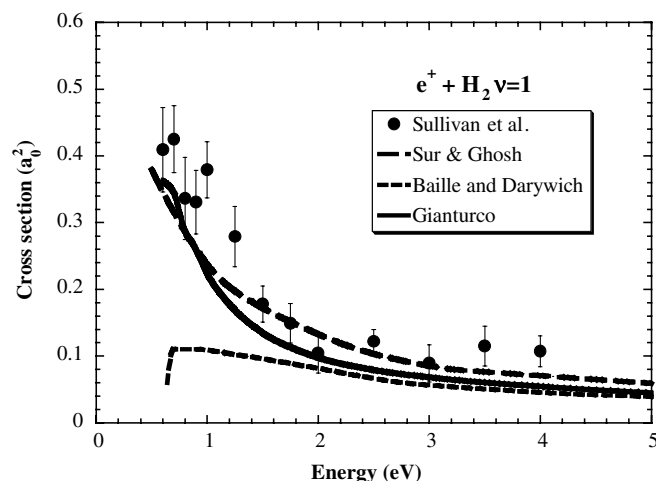


Fig. 10. Vibrational excitation of H<sub>2</sub> by positron impact. The experimental results [48] are compared with three theoretical approaches [50–52].

system can be met, but the signs are encouraging for single channel excitation cross-sections.

## 5. Conclusions

For electron scattering there are now a significant number of what might be considered benchmark systems, where experiment, and experiment and theory agree at the 5–10% level across a number of incident energies and scattering angles, for a number of different processes. Some significant challenges remain however, particularly in the case of polyatomic molecules, and particularly for theory. In contrast there are few systems in positron scattering where it can be said that benchmarks have been established. However, there are promising new developments in the field, particularly with the development of new, high resolution beam sources. It is hoped that this will encourage further theoretical treatments of the positron scattering process, especially leading towards a theoretical treatment of positronium formation. It can be reasonably hoped that benchmarks for positron scattering will be established within the next decade, at least for simple systems, for a variety of positron interaction processes.

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