COLLISIONALLY EXCITED FEW-ELECTRON SYSTEMS: THEORETICAL INTRODUCTION AND SURVEY

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ABSTRACT

We consider excitation, ionization, and charge transfer in collisions of protons (and antiprotons) with the single-electron targets H, He^+, and Li^{2+}. These collisions are first compared to other types of ion-atom collisions. A brief review of our own theoretical method is given; in particular we describe how we allow for both large charge transfer and ionization probabilities while retaining the computational efficiency that allows us to consider a variety of collision partners and collision energies. We comment on the comparison of our results to other theoretical work and to experiment. The qualitative features of the various inelastic cross sections are discussed, in particular how they scale with collision energy, target nuclear charge, and the sign of the projectile charge.

INTRODUCTION

There are many reasons why theoretical and experimental studies of atomic collisions are important. Aside from the technological applications of atomic collisions and the role they play in other branches of physics, atomic collisions provide a means for learning about collision physics itself. In this regard atomic collisions have a distinct advantage over nuclear and elementary particle collisions, in that for the atomic case the fundamental two-body (nonrelativistic) force is known. Furthermore, due to the strong forces between the electrons and atomic nuclei the independent particle model (IPM) is always a reasonable starting point and is often (particularly for inner-shell processes) an excellent approximation. For bare ion projectiles, the projectile is clearly distinguishable from the target electrons whose excitation, ionization, and capture are being studied. For all but very low impact velocities the bare ion projectile accurately can be assumed to be moving on a classical path. An accurate description of the electronic structures of the separated projectile and target is usually not difficult to obtain. With all this the dynamical collision

problem can be reduced to one for the individual electrons.

Despite these simplifying features, there is a great richness of phenomena in atomic collisions. As this paper will attempt to emphasize, the collision parameters (collision velocity, target Z, and projectile Z) can be, and have been, varied over wide ranges. There are several quite distinct types of collisions, each with its own dominant processes and important physics.

This is a conference on atomic inner shell physics. One subclass of collisions involving inner shell electrons is K- and L-shell excitation, ionization, and charge transfer in collisions of small bare ions (nuclear charge $Z_p \leq 3$ or so) with neutral target atoms of nuclear charge Z_T greater than 10 or so. Inner-shell inelastic processes are important for collision energies that give velocity matching between the projectile and the target electrons in the shells in question. For inner shell electrons the electron-nucleus force dominates, and the IPM is an excellent approximation. The description of the target electronic structure is thus relatively simple, and one can concentrate on the collision physics. For these collisions ionization is by far the dominant inelastic process; excitation and charge capture probabilities are quite small. The most widely used theoretical model, and one which is quite successful in describing many features of the collisions, is the increased binding correction of the plane wave Born approximation (PWBA) that was developed some 10 years ago by Brandt and co-workers.[1] Our own contributions involve coupled-channel calculations with a single-center basis.[2] Discrete pseudostates are used to span the ionization continuum. Projectile centered functions are not needed in the expansion of the time-dependent single electron orbitals of the collision system because the flux in the charge transfer channels is very small. We have shown that the small charge transfer probability can be computed perturbatively from our single-centered expansion.[3] In asymmetric collisions $(Z_p \ll Z_T)$ an important electron capture mechanism is a two-step process in which the electron to be captured is first excited to a target continuum state which is resonant in energy with the state on the moving projectile into which the capture is to occur. The width of the energy resonance of this two-step process is very narrow when $Z_p \ll Z_T$, so care must be exercised in the manner in which the exact continuum spectrum of the target hamiltonian is replaced by a discrete pseudostate representation.[4,5]

This paper is however part of a symposium on collisional excitation of few-electron systems, and the remainder of the paper will deal specifically with single-electron collision systems rather than inner shells of neutral atoms. The inner shell case has the complications (and therefore interesting physics!) of a many-fermion system. Even though the independent particle model may be accurate, the Pauli exclusion principle imposes a correlation among the electrons.[6] Consider K-shell vacancy production as a simple example. One mechanism is direct ionization of a K-shell electron. But there are others. For example, in a single collision an

L-shell electron could be ionized and then the K-shell vacancy made by excitation of a K-shell electron into the L-shell hole that was just made. On the other hand, a K-shell vacancy is not made when a K-shell electron is ionized but then in the same collision an L- or M-shell electron is deexcited into the K-hole. All such processes must at least formally be considered,[6] and in some cases are important in practice.[3,7] For multi-electron targets there is also the possibility of breakdown of the IPM. Rearrangement of a number of the electrons during the course of a collision can cause the effective single electron potentials to vary in time and to be different in each multi-electron channel.

Single-electron collision systems avoid these complications. There has been to our knowledge no experimental work on single-electron collision systems in the $Z_p \ll Z_T$ asymmetric region in which our inner-shell studies have been carried out. Such experiments would be very useful in testing our understanding of this region of collision parameters. There has been a lot of effort, both experimental and theoretical, in the inverse region where $Z_p \gg Z_N$, i.e. highly stripped or bare heavy ions in collision with atomic hydrogen. One characterizing feature of these collisions is the importance, particularly at lower collision energies and for large ionic charge q of the projectile, of capture into a large number of highly excited orbitals on the heavy projectile. Due to the large number of discrete states involved, standard coupled-channel calculations with all important channels included can be prohibitively expensive. We have done some computational work on $He^{2+}+H(1s)$ and $Li^{2+,3+}+H(1s)$ collisions,[8,9] but will have nothing further to say here about such collisions.

The rest of this paper deals with single-electron collisions for which $Z_p \lesssim Z_N$, specifically p+H, p+He$^+$, and p+Li^{2+}. As we will show, the ratio of capture to ionization cross sections varies over a wide range as the collision velocity is changed. Excitation cross sections, for example to the 2s and 2p states, are the same order of magnitude as the ionization, and are included in the discussion. Our method, particularly how it accounts for the large charge transfer flux that can occur in these collisions, is very briefly reviewed in the next section. We then discuss our results. We briefly outline the comparison between our results and those of experiment and other theories. But the emphasis is on the qualitative features of the cross section results, and in particular how they scale among the various sets of collision partners.

THEORETICAL METHOD

The method we used for the calculations being discussed here has been extensively described in the literature,[10] and here we only briefly review some of the most important features. We are considering collisions for which the projectile velocity is approximately (to within a factor of 10) equal to the velocity of the

target electron. For heavy projectiles (specifically protons) the projectile motion can be treated as classical. In all our calculations discussed here the projectile is taken to move with constant speed and in a straightline.

Coupled-channel methods use a basis set expansion of the time-dependent wave function $\Psi_\lambda(\vec{r},t)$ of the electron in the collision system. The target-centered expansion functions we use are obtained by diagonalizing the target hamiltonian H_T in underlying finite basis sets of square integrable functions. The pseudostates for which the eigenvalues of the projected hamiltonian lie above the ionization threshold provide a discrete representation of the ionization continuum. Examination of the spectrum of eigenvalues from the diagonalization of H has proven to be a very useful assessment of the adequacy of the basis sets being used. For example, certain basis functions can lead to pseudostates of such high energy that they do not participate in the dynamics of the collision. Adding such functions to the basis without prediagonalization of H_T would lead to a false sense of convergence.

The conventional two-center expansion (TCE) used by others employs an expansion of the form

$$\Psi_\lambda \text{ (TCE)} = \sum_{n=0}^{N} a_{n\lambda}(t)\chi_n(\vec{r},t) + \sum_{m=0}^{M} b_{m\lambda}(t)\phi_m(\vec{r},t). \qquad (1)$$

The functions χ_n and ϕ_m are target and projectile centered, respectively, and diagonalize the respective hamiltonians. This expansion has the defect that with it solving the coupled equations for the expansion coefficients is very time consuming. The problem arises from the fact that for finite times the χ_n and ϕ_m are not orthogonal, and that for large N and M the expansion leads to a large number of the difficult exchange matrix elements. The TCE also has the at least formal defect that for finite t the expansion is overcomplete. For finite t and large N the ϕ_m have unit projection onto the set χ_n, and the equations for determining $a_{n\lambda}$ and $b_{m\lambda}$ must become ill-conditioned. This overcompleteness also points up the lack of economy in the TCE: why include the ϕ_m for those times where they add nothing to the expansion?

Our first coupled-channel calculations were for asymmetric $Z_p \ll Z_N$ collisions, in which the charge transfer flux comprises a very small part of the wave function Ψ_λ. For calculations of ionization the projectile centered part of the wavefunction is hence unnecessary, and the charge transfer probability can be calculated with a t-matrix expression.[4] We thus used a single-center expansion

$$\Psi_\lambda(SCE) = \sum_{n=0}^{N} a_{n\lambda}(t) \, \chi_n(\vec{r},t) \qquad (2)$$

and found it to be not only very efficient but also accurate.

For the collisions we are considering in the present paper though, for which $Z_p \sim Z_N$, the charge transfer probability can be large and the single center expansion is not an efficient way to proceed. To retain the computational speed of the SCE but still allow for the charge transfer flux we invented[10] what we call the 'one and a half centered expansion', in which

$$\Psi_\lambda(OHCE) = \sum_{n=0}^{N} a_{n\lambda}(t)\chi_n + \sum_{m=0}^{M} b_{m\lambda}(\infty)\beta_m(t)\phi_m. \qquad (3)$$

The OHCE differs from the TCE in that the OHCE prechooses $\beta_m(t)$ as a fixed function. The choice made for $\beta_m(t)$ is constrained in principle only by the boundary conditions that $\beta_m(-\infty)=0$ and $\beta_m(\infty)=1$, so that the $b_{m\lambda}(\infty)$ are charge transfer amplitudes. The time dependent expansion coefficients $a_{n\lambda}(t)$ and time independent ones $b_{m\lambda}(\infty)$ are determined from applying the conditions

$$\left\langle \chi_n \left| i\hbar \frac{\partial}{\partial t} -H \right| \Psi_\lambda(OHCE) \right\rangle = 0 \qquad (4)$$

and the auxiliary constraint

$$0 = \int_{-\infty}^{\infty} \langle \phi_m | i\hbar \frac{\partial}{\partial t} -H | \Psi_\lambda(OHCE) \rangle dt. \qquad (5)$$

With this specific choice of auxiliary constraint we have what we have called the perturbative version of the OHCE (POHCE), and it is the only version of the method that we consider here. If $\Psi_\lambda(OHCE)$ is replaced by $\Psi_\lambda(SCE)$ in eq. (4) we recover the method we used previously for asymmetric collisions where charge transfer is small; hence the name perturbative. All calculations described here take $\beta_m(t)$ to be a unit step function at $t=0$ and retain only a single state, the 1s, in the projectile-centered part of the expansion in eq. (3).

Our expansion then is characterized by a large number of target-centered functions (which we are able to use because our method is comparatively so much more efficient than the TCE), but also with allowance for flux loss in the dominant charge transfer channel. The large target-centered expansion allows the ionization of the target to be well represented. A necessary, but not sufficient, test of the adequacy of our representation of the ionization

continuum is that in the limit of small Z_p, where our calculation reduces to the semiclassical first Born approximation for ionization, we accurately reproduce the plane wave Born ionization calculated with exact target continuum wavefunctions.

RESULTS AND DISCUSSION

We have applied the POHCE to collisions of protons with ground state H, He$^+$, and Li^{2+}. Comparison of our cross sections to experiment and other calculations has been given elsewhere,[8-10] so we only make a few brief remarks here. For p+H and collision energy E$\geq$15 keV the POHCE was found to give very good agreement with experiment and with extensive TCE calculations of Shakeshaft[11] for n=2 and n=3 excitation, ionization, and charge transfer. An example is given in fig. 1, where our ionization cross section is compared to that of Shakeshaft and to the very recent experiment of Shah and Gilbody.[12] The agreement among the three results, while not perfect, is overall quite good. We note that we used only s, p, and d angular momentum states in our target-centered expansion,
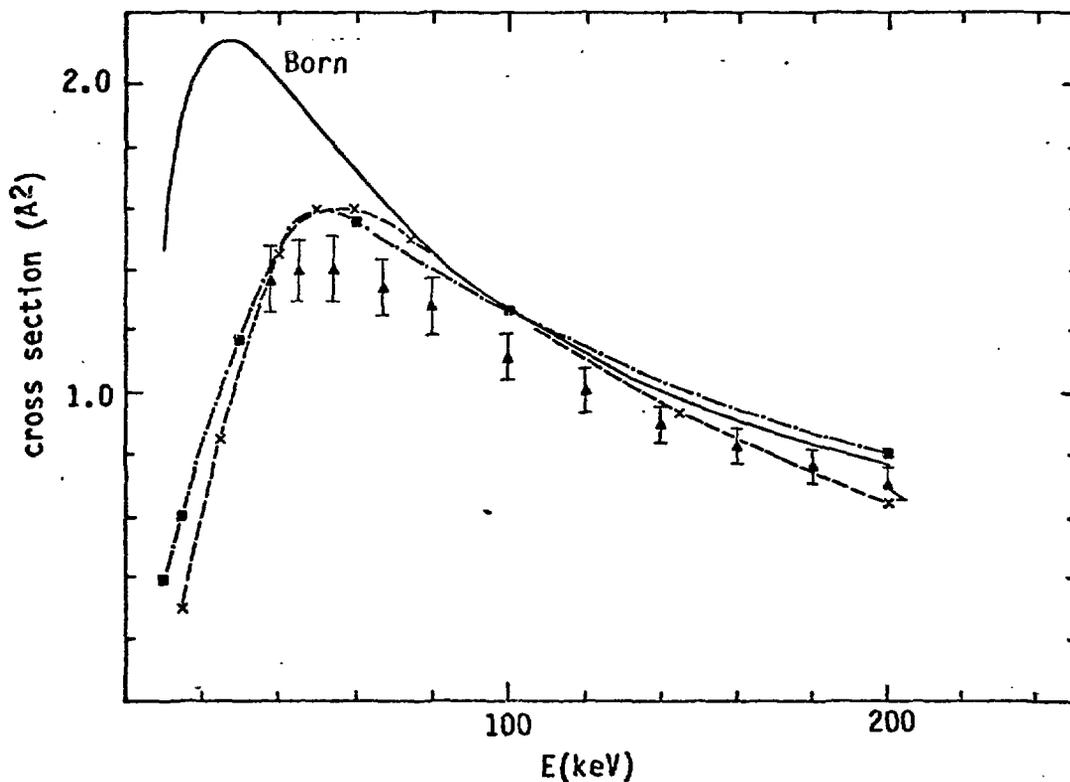

Fig. 1. Ionization cross section for p+H collisions. The experimental points are from Shah and Gilbody[12]. The 3.8% uncertainty in overall normalization has been included in the error bars. The squares, connected by a chain curve as a guide to the eye, are our POHCE calculation, and the crosses, connected by the dashed curve, are from the TCE calculation of Shakeshaft[11].

so from the coupled equations calculation we have only the ioniza-
tion in these partial waves. To approximately include the higher
partial waves, we added their contribution as given in first Born.
It is difficult to see how to make a similar correction in the TCE,
and that may partly account for why our cross section at 200 keV
lies above that of Shakeshaft.

For p+He[+] the situation is not as good.[8] An example is
given in fig. 2, where our POHCE capture cross section is compared
to experiment[13] and to the calculations of Winter,[14] the most
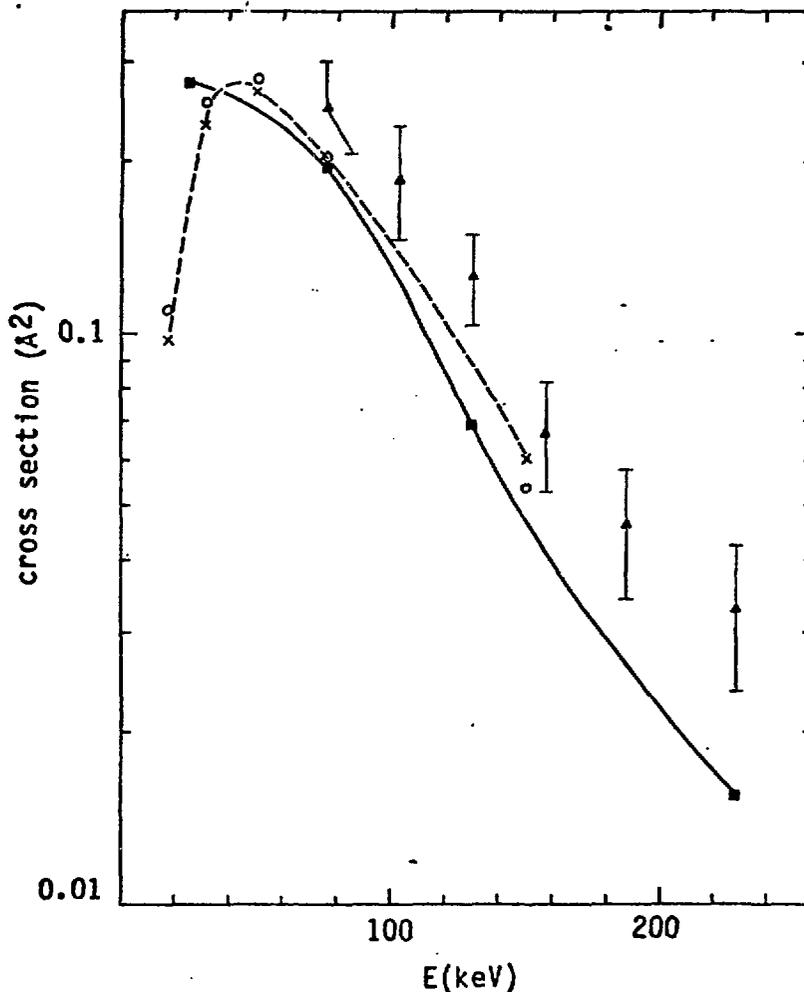extensive TCE calculations published so far for this system. We

Fig. 2. Capture cross section for p+He[+] collisions. The experi-
mental points are from Angel et al[13]. The 13.6% uncertainty in
absolute magnitude has been included in the error bars. The
squares connected by a solid line to guide the eye are our POHCE
calculations, where we calculate only capture into H(1s) and assume
excited state capture is 20% of this. The crosses connected by a
dashed curve, are from the TCE calculation of Winter[14]. The open
circles are 1.2 times Winter's ground state capture.

have included the 13.6% uncertainty in the absolute magnitude of the experimental results in the error bars. We calculate only capture into H(1s) and assume that excited state capture is the 20% of this that one gets by assuming n^{-3} scaling. The agreement between our calculation and that of Winter is only fair. Part of the discrepancy is that his excited state capture differs from our assumption of 20% of ground state capture. We are significantly below the experiment, particularly at the higher energies. It is interesting that our total electron loss cross section (capture plus ionization) is in rather good agreement with experiment. Apparently either our calculation or the experiment fails to distinguish properly between ionization and charge capture.

For $p+Li^{2+}$ we know of no experimental data with which to compare. There are data for $p+Li^{+}$, and we have carried out calculations for this two-electron system.[9] The comparison between our calculation and experiment is overall similar to what it is for $p+He^{+}$, except that in addition the total loss cross section we compute is somewhat below experiment.

We now discuss the overall qualitative features of the cross sections we have computed for these systems, particularly their energy dependence and scaling in the target nuclear charge Z_T. The 1s to 1s capture, σ_{KK}, decreases sharply with collision energy E, and at a given E/Z_T^2 it scales approximately as Z_T^{-5}. The ionization cross section σ_I is fairly flat in E over the same collision energy range and scales approximately as Z_T^{-4} (the Z_T scaling in the first Born). Hence the ratio of capture to ionization, σ_{KK}/σ_I, scales roughly as Z_T^{-1} and falls sharply with E in the energy range we are considering. This is shown in fig. 3. As Z_T and E are varied in the collisions we have considered, the ratio of capture to ionization changes by over two orders of magnitude.

For asymmetric collisions ($Z_p \ll Z_T$) the excitation and ionization cross sections are accurately given by the first two terms in the Born expansion, $\sigma = \sigma_B(1+f\rho)$, where $f = Z_p/Z_T$ and ρ is independent of Z_T and Z_p. The Born cross section σ_B is proportional to Z_p^2, so the correction to the Born scales like Z_p^3. For $Z_p > 0$, at low energies ρ is negative and has been associated physically with the increased binding of the target electron when the projectile is well inside the electron's orbital radius[1]. A novel way we have illustrated this effect is by performing coupled-channel calculations with negative Z_p[15]. For low energy $p^{\pm}+Cu$ collisions then the cor-
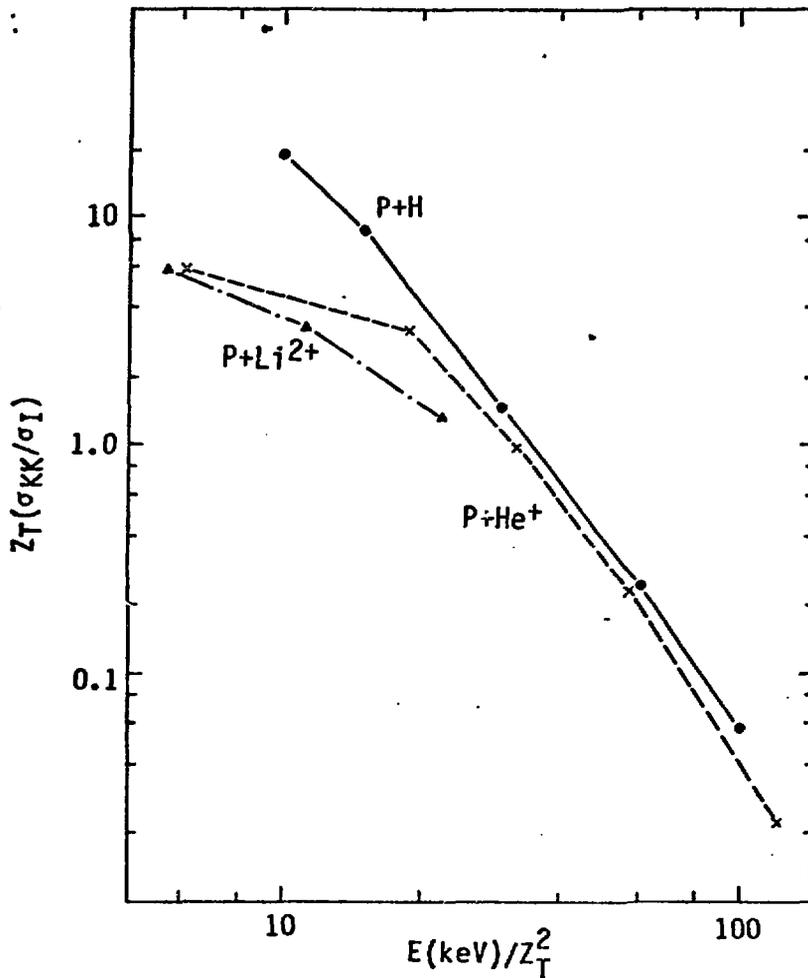
Fig. 3. The ratio of capture to ionization cross sections, multiplied times Z_T and plotted as a function of scaled collision energy for the indicated collision partners.

rection $Z_p \rho$ is negative for $Z = +1$ (increased binding) and positive for $Z_p = -1$ (decreased binding). In figs. 4-6 the behavior of the ratio $R = \sigma/\sigma_B$ is followed into the higher scaled collision energy and more symmetric collision regime. The $Z_p = -1$ results for Cu connect smoothly, in a qualitative sense, to the results for smaller Z_T. There is a change in the magnitude of the Z_p^3 correc-
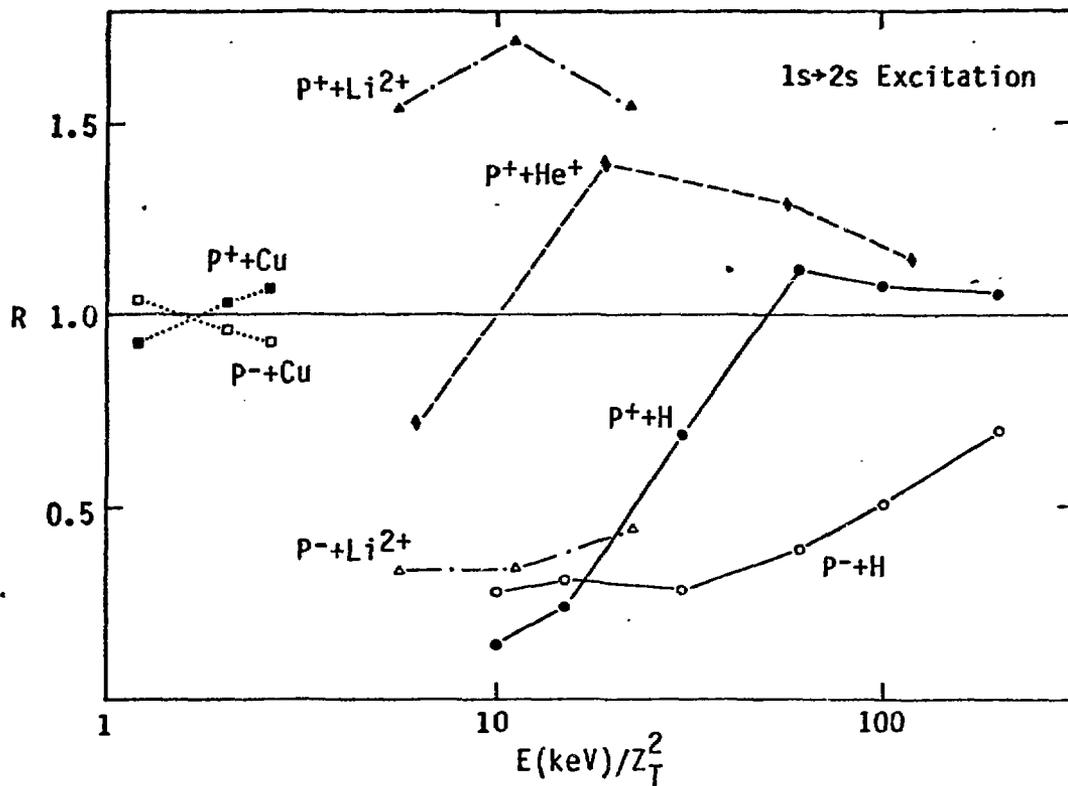
Fig. 4. The ratio R of the POHCE 1s→2s excitation cross section to the first Born, plotted as a function of scaled collision energy for the indicated collision partners. The copper calculations were done using the Hartree-Fock potential of the neutral atom.

tion as Z_T is decreased by over a factor of ten in going from the highly asymmetric collision to the more symmetric ones. In each case (excitation to 2s and 2p and ionization) there is 'decreased binding' (R>1) at low energies and a transition to 'antipolarization' (R<1) as E is increased. The Born correction does not scale as f for the small Z_T cases, but at least does decrease as Z_T increases.

The behavior for $Z_p=+1$ is on the other hand quite different. For $p^{\pm}+Cu$ the p^+ and p^- cross sections are symmetrically placed on opposite sides of the Born, but for $Z_p \sim Z_T$ that is the case only
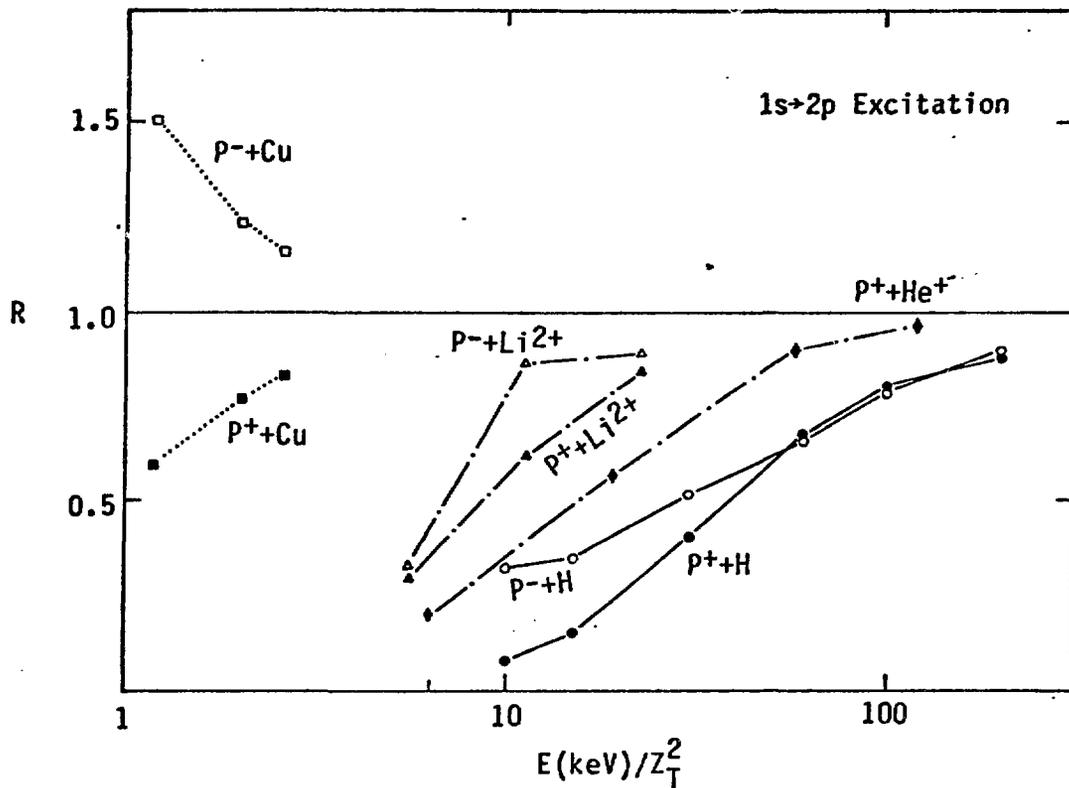
Fig. 5. As in fig. 4, but for 1s→2p excitation.

at high energy. We ascribe the qualitative difference in the be-
havior of R for Z_p=+1 to be due to the existence of a charge trans-
fer channel for Z_p=+1 but its complete absence for Z_p=-1. It ap-
pears that when charge transfer is large, it robs flux from both
the excitation and ionization channels. For example, at $E/Z_p^2 \sim 10$
keV charge transfer becomes very large as Z_T is decreased. Corre-
spondingly, R is sharply depressed below the value one would extra-
polate from p$^+$+Cu when one goes from Li^{2+} to He$^+$ to H targets.

Another striking feature of the results shown in figs. 4-6 is
the very close agreement above E~50 keV between the 2p excitation
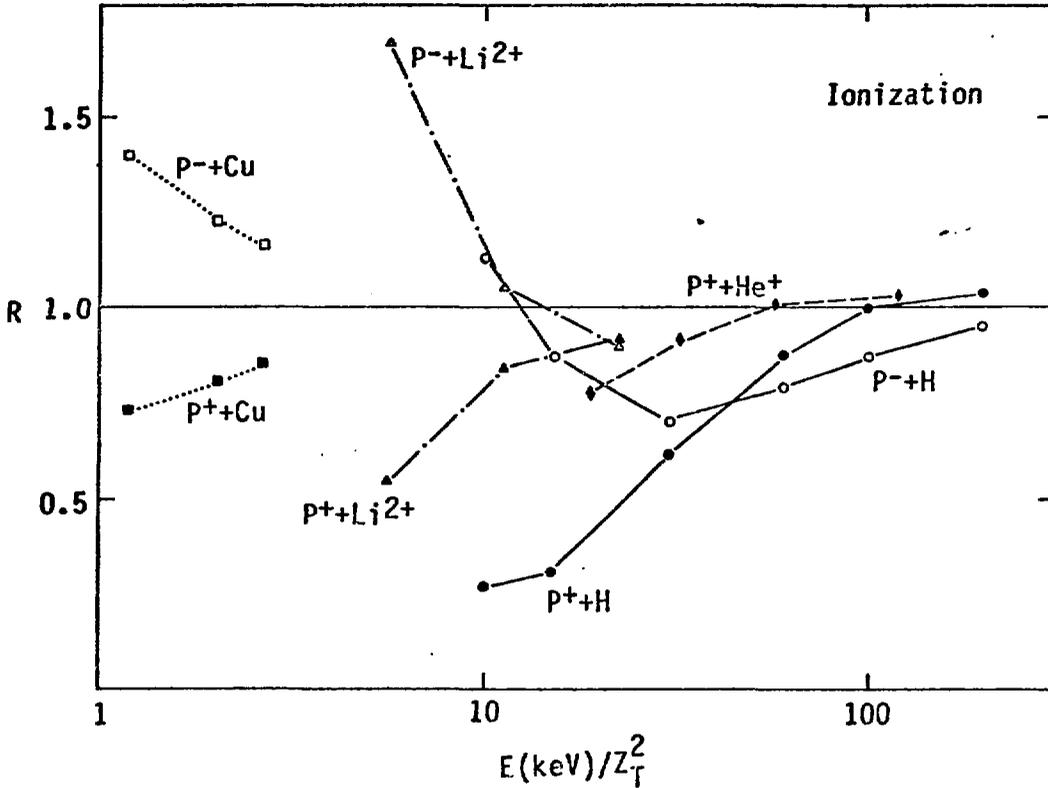cross sections for p$^+$+H and p$^-$+ H, and to a lesser extent for the

Fig. 6. As in fig. 5, but for ionization.

ionization. We do not presently understand the physical origin of this effect, and feel in fact that there is much still to be learned from results of the sort presented in these figures. We note that it would be very useful to have experimental results for 2s and 2p excitation in the p+He$^+$ and Li^{2+} cases, so that our Z_T scaling of these excitation cross sections could be tested.

In conclusion, collisions of bare ions of Z =±1 with one-electron targets of small nuclear charge Z_T constitue one distinct type of ion-atom collision. Such collisions are characterized by large collision strength $f=Z_p/Z_T$ and large charge transfer. Our OHCE method allows us to consider a variety of values for the collision parameters. We find qualitative differences in the collision physics here compared to what obtains in the $Z_p \ll Z_T$ asymmetric

region that is most often explored in inner-shell collision studies. We plan further study of these Z_p-Z_T collisions by extending our OHCE methods to system of several electrons where IPM breakdown is extensive.

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