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ELECTRON-IMPACT EXCITATION OF IONS

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1. INTRODUCTION

The ancient curiosity as to "what produces the light" still motivates research on electron impact excitation of ions. We have developed extensive knowledge of the subject which rests on detailed information about atomic structure and a conviction that quantum theory, in principle, correctly describes the atomic structure and the collision dynamics. Still, we are unable to predict cross sections or rates for this process with sufficient reliability to satisfy our needs.

In astronomy and astrophysics, observations from spacecraft allow detection of light (from ions) without modification by the Earth's atmosphere so that considerably more detailed data are available and correspondingly a more accurate understanding of the underlying basic processes is needed. In the development of a controlled fusion energy source, the electron impact excitation of ions provides unique information on the conditions in the plasma and can be a decisive factor in the energy balance affecting plasma evolution. A wide variety of investigations in spectroscopy, plasma properties, and basic collision studies require known cross sections for excitation of ions.

The electron impact excitation of ions has several unique aspects. The light emitted during relaxation of excited ions is generally of shorter wavelength than light from other processes. The coupling of excitation with autoionization is relatively more important for ions

than for atoms. Ion excitation cross sections are finite and often largest at the threshold energy. At the same time the calculations are most difficult near threshold. The processes of excitation, ionization, and recombination are so strongly interrelated in electron-ion collisions that independent treatments of these problems may not be sufficient.

In order to study electron impact excitation of ions under sufficient control to determine specific cross sections as a function of collision energy, it is generally necessary to produce ion beams of specific species, charge, and atomic state and to interact the ion beam with a controlled electron beam. Since attainable ion beam densities are tenuous (typical beams of 10^5 particles/cm³ compare to 10^{-11} torr pressure), few definitive experiments have been accomplished. Attempts to study electron impact excitation of ions without recourse to controlled ion beams are typically unable to produce sufficiently detailed data to test understanding of the basic process.

This paper will review electron-ion beams experiments primarily. The techniques, difficulties, and present trends in this area will be discussed. Measured cross sections will be compared with theoretical results and the current level of agreement assessed.

2. THEORY

2.1 General

The theory of electron impact excitation of ions has been recently reviewed by Henry;¹ it was discussed by Robb² at the preceeding NATO summer institute; and it has previously been reviewed by several others.³⁻⁵ There are a great number of individual predictions of excitation for particular transitions of specific ions. Henry¹ lists over

350 references, the Los Alamos compilation⁶ gives comparative results from more than one calculation for each of more than 100 transitions, and the current ORNL compilation on excitation of iron ions⁷ lists calculated cross sections for roughly 800 transitions. This vigorous activity in excitation theory has produced some concensus as to which approximations are most reliable in spite of the paucity of experimental results which are needed to serve as a standard to test the accuracy of the various theoretical approaches. The discussion which follows makes no attempt to provide equations which would allow actual calculations, but examines standard approaches for physical insight.

The accepted non-relativistic theoretical approaches all begin with the Schroedinger equation for the system of $N + 1$ electrons:

$$H(Z, N+1)\psi = E\psi \quad (1)$$

where N and Z are the number of electrons and nuclear charge of the ion, ψ is the total wave function of the $N + 1$ electron system, and E is the total energy, $E = E_i(Z, N) + k_i^2$ (which defines the kinetic energy, k_i^2 , of the incident electron in Rydbergs).

The Hamiltonian is

$$H(Z, N+1) = - \sum_{i=1}^{N+1} \left(\nabla_i^2 + \frac{2Z}{r_i} \right) + \sum_{j=i+1}^{N+1} \sum_{i=1}^N \frac{2}{r_{ij}} \quad (2)$$

Having written these standard expressions we immediately encounter difficulty because Eq. (1) is a many-body problem, never less than three bodies. In principle there is no compromise in expanding the many-body wave function in terms of products of single-particle wave functions:

$$\Psi(N+1) = A \sum_i \theta_i(N+1) \chi_i(X_1 \dots X_N) \quad (3)$$

where A antisymmetrizes the total function and X_n denotes coordinates of the n th electron. Substitution of Eq. (3) into Eq. (1) results in a number of coupled differential equations. Generally expansion (3) is chosen so that the functions χ_i are the solutions of the next simpler problem (i.e., they are the eigenvectors of the N electron ion which is the target), and the $\theta_i(N+1)$ are coefficients but they also are the wave function of the free or incident electron. The wave functions for the bound-state system, χ_i , are usually antisymmetrized products of one-electron functions determined from the solution of a central field model such as the Hartree-Fock Method. It should be further noted that the index, i , stands for all the system quantum numbers including parity, angular momentum, and spin. Thus, Ψ is a sum over coupled, antisymmetrized, product wave functions which leads to messy algebra. One additional modification to Eq. (3) is usually specified:

$$\Psi(N+1) = A \sum_i \chi_i \epsilon_i + \sum_j C_j \phi_j(X_1 \dots X_{N+1}) \quad (4)$$

where the C_j are numerical coefficients and ϕ_j are bound-state wave functions of the $N+1$ electron ion or atom of next lower charge. The addition of the second term in Eq. 4 is needed to provide a more complete set mathematically,¹ but note that the ϕ_j states are important physical quantities in this problem as well. Some of these states are schematically represented for an arbitrary Li-like ion in Fig. 1. Of the eigenstates represented in Fig. 1, only those below the $1s^2 ({}^1S)k$ continuum and the lowest of the levels illustrated below the $1s2s({}^3S)k$ continuum are fully bound states. The other discrete levels illustrated are autoionizing

resonances. The arrow at the far left in Fig. 1 illustrates a particular total energy in a scattering of a free electron by the He-like ion. For such total energies an autoionizing resonance of the Li-like ion may have coincident energies, and near this condition the resonances influence the excitation cross sections of the He-like ion (see Ref. 8 for a recent calculation including such resonances).

2.2. Coupled State Theory

At any rate, if expansion (4) is substituted into Eq. (1) and projected onto the angular momentum components of a particular channel, i , we obtain the close-coupling equations which are of the form:

$$-(\nabla^2 + k_i^2)\theta_i(X) + \sum_f V_{if}(X)\theta_f(X) = 0 \quad (5)$$

where X represents the space and spin coordinates and

$$V_{if}(X_1) = \frac{-2Z}{r_1} \delta_{if} + \int \psi_1(X_2 \dots X_{N+1}) \sum_{n=2}^{N+1} \frac{2}{r_{1,n}} \psi_f(X_2 \dots X_{N+1}) dX_2 \dots dX_{N+1} \quad (6)$$

The asymptotic form of this potential is

$$V_{if}(X) \underset{r \rightarrow \infty}{\sim} \frac{-2q}{r} \delta_{if}$$

where $q = Z - N$ is the ionic charge; but note that f is an index here which runs over all of the states included in the expansion (4). In seeking solutions to the set of equations (5), various approximations are made.

Assuming that the continuum electron moves in a central field,

the form of θ_i can be:

$$\theta_i(X) = Y_{\ell m}(r) \delta(\text{spin}) \frac{1}{r} F_i(r)$$

where the $Y_{\ell m}$ are spherical harmonics and the radial components $F_i(r)$ which satisfy Eq. (5) will have asymptotic form:

$$F_i(r) \underset{r \rightarrow \infty}{\sim} \frac{1}{\sqrt{k_i}} \{ \sin(\zeta_i) \delta_{if} + \cos(\zeta_i) \rho_{if} \} \quad (7)$$

where $\zeta_i = k_i r - \frac{1}{2} \lambda_1 \pi + \frac{q}{k_i} \ln(2k_i r) + \arg \Gamma(\lambda + 1 - \frac{iq}{k_i})$ is the asymptotic phase of the regular Coulomb function. Expression (7) has included in it the quantity we ultimately want. The term $\sin(\zeta_i) \delta_{if}$ represents elastic scattering while the term $\cos(\zeta_i) \rho_{if}$ represents inelastic scattering state i to state f . The ρ_{if} is an element of a matrix, related to the reactance or R-matrix, which has properties analogous to the more familiar scattering amplitude $f(\theta)$ used in the partial wave approach to simple elastic scattering problems (see Ref. 9 for example).

The matrix element ρ_{if} can be obtained by use of the Kohn variational principle:

$$\delta \{ \langle \psi_i | H - E | \psi_f \rangle - c_{if} \} = 0 \quad (8)$$

Recall that f is still an index which runs over all states included, so that the number of equations must be limited, that is the number of functions (4) is truncated at I so that the index i (or f) runs from 1 to I . An I -state close-coupling approximation then results. The cross section for excitation of a channel i to channel f is ultimately represented qualitatively by

$$\sigma_{if} \propto |c_{if}|^2 \quad (9)$$

Actual solutions by computer (for instance using the code RMATRX¹⁰) involves computation of a set of one-electron basis functions, considerable vector-coupling algebra, and many matrix inversions. Solutions for simple ions and a few states I are tractable but complex ions and/or many states, I , are generally not attempted.

2.3. Distorted Wave and Coulomb-Born Approximations

The difficulty with Eq. (5) is that H is coupled through the V_{if} ($i \neq f$) part of the potential in Eq. (6). A substantial simplification results from uncoupling the equation which can be accomplished by setting $V_{if} = \begin{cases} 0 & \text{if } i \neq f \\ V_{ii} & \text{if } i = f \end{cases}$ where V_{ii} is given by Eq. (6) with $f = i$. The differential Eq. (5) now has only one term in what was the summation, and there is only one index, i , left in Eq. (5) so that each differential equation stands alone. Now when the variational principle [Eq. (8)] is applied to find the matrix ρ_{if} , the state f is one particular, selected, final state, and calculation of excitation to any selected final state is a two-state problem. This is the distorted wave approximation. Coupling of states by the incident electron has been lost but the technique is sufficiently simple to apply to quite complex ions. Of course the actual approximation of Eqs. (5) and (6) can be done many ways. Henry¹ distinguishes about ten variations of distorted wave in published work.

The Coulomb-Born approximation is a simple modification of the potential (6) which provides additional simplification. The asymptotic form of Eq. (6) $V_{if} = \frac{-2q}{r} \epsilon_{if}$ is used for the problem at all r . Note that the first term of Eq. (6) is not the same as this in that the nuclear charge Z has been replaced by the ionic charge, q . The screening of the bound electrons which was determined by the integral part of Eq. (6)

in a flexible (r dependent) manner even in distorted wave is now fixed to be simply full screening at any r . The plane-wave Born approximation takes $V_{if} = 0$ which would correspond to $q = 0$ in the Coulomb-Born approach and is inappropriate for ions.

Other approximations can be applied to any of the three methods mentioned. Unitarity has a specific mathematical meaning and should be applied to the matrices derived from the ρ_{if} . Physically unitarity corresponds to conservation of flux in that the probabilities of all allowed scattering must sum to the initial flux. Naively it would seem that unitarity should always be satisfied but, if the elements of ρ_{if} are all small, it may not be essential to obtaining a good approximation.

Exchange has not been specifically mentioned. In principle, if all the wave functions in the expansion (4) and the potential (6) are fully antisymmetrized, exchange is explicitly included throughout the problem but with a great addition in the real work. Thus numerous different approximations are in fact made in the way and extent to which exchange is included.

At this point it is instructive to glance ahead to Fig. 5 for excitation of Be^+ . All of the various approximations discussed have been applied and convergence of the approximations toward the experimental cross section reflects the quality of the approximations as inferred above.

2.4. Oscillator Strengths, Gaunt Factors, and Collision Strengths

If the general theoretical approach outlined above is followed but for only bound states, that is with trial wave functions ψ which have no continuum electron function and which consequently obey $\psi \rightarrow 0$ as $r \rightarrow \infty$, then a related but much simpler problem is solved. Expression (8) results

in oscillator strength rather than a reactance matrix. Seaton¹¹ and Van Regemorter¹² wrote down a specific expression for connection between excitation cross sections and oscillator strengths:

$$\sigma_{if} = \frac{8\pi}{\sqrt{3}} \frac{f_{if} \bar{g}}{E \Delta E_{if}} \pi a_0^2 \quad (10)$$

where E ($= k_i^2$ earlier) is the incident electron kinetic energy (in Rydbergs), ΔE_{if} is the transition energy (in Rydbergs), f_{if} is the oscillator strength, and $\bar{g}$ is called the effective Gaunt factor which serves to adjust Eq. (10) in a systematic manner based on knowledge of ratios of cross sections to oscillator strengths. When large numbers of excitation cross sections are required, for example in plasma modeling,¹³ Eq. (10) is generally used.

From early comparisons $\bar{g}$ was taken to be 0.2 near threshold with an explicit functional variation at higher energies. From an experimental viewpoint, Taylor et al.¹⁴ found that for Ca^+ , Ba^+ , Hg^+ , and N_2^+ measured cross sections, this value of $\bar{g}$ agreed with experiment to within a factor of two as originally advertised. For the simple $\text{Be}^+(2s - 2p)$ case the $\bar{g}$ formula with $\bar{g} = 0.2$ at threshold fits the experimental data¹⁵ as well as any theoretical result. However, for ions of charge greater than +1, taking $\bar{g} = 0.2$ seriously underestimates cross sections obtained from better theoretical approaches or the only experimental cases available.¹⁶ Bely¹⁷ and Blaha et al.¹⁸ had noted earlier that for multiply charged ions this value of $\bar{g}$ was probably too small. The most recent examination of the $\bar{g}$ approximation by Younger and Wiese¹⁹ found that for multicharged ions and $\Delta n = 0$ transitions, $\bar{g}$ near 1.0 was best. For $\Delta n \neq 0$ and for optically forbidden transitions they found $\bar{g}$ varies from 0.05 to 0.7

for conditions appropriate to plasma modeling applications. The $\bar{g}$ predictions are not sufficiently reliable in many situations.

It is common in all theoretical predictions of excitation to present results as collision strengths, Ω_{if} . This quantity is the particle collision equivalent of the oscillator strength. The connection between collision strength and cross section is always

$$\Omega_{if} = w_i E \sigma_{if} \quad (11)$$

where E is again the incident electron kinetic energy in Rydbergs, w_i is the statistical weight of the lower level [$w_i = (2S_i + 1)(2L_i + 1)$ or $(2J_i + 1)$ if fine structure is taken into account, where S_i , L_i , and J_i are the usual total angular momentum quantum numbers of the state i], and σ_{if} is the cross section in units of πa_0^2 .

3. EXPERIMENTAL TECHNIQUES

3.1. Plasma-Based Rate Measurements

This paper will deal only superficially with excitation rate measurements, but it is worth noting that a few detailed studies have been performed. The 1972 review of Kunze²⁰ remains the standard reference for describing measurement techniques, and Gabriel and Jordan²¹ have given a review of the interpretation for spectral intensities from plasmas. The intensity, I_{fj} , of radiation of a given transition from a well-characterized plasma, per unit volume, per steradian can be written

$$I_{fj}(t) = \frac{h\nu}{4\pi} \frac{A_{fj}}{\sum_r A_{fr}} N_e(t) N_i(t) \alpha_{if} \quad (12)$$

where N_e is the electron density, N_i is the ion density in the initial state, α_{if} is the rate coefficient for excitation from state i to f , A_{fj} is the transition probability for the observed radiation from state f to a specific state j , and A_{fr} is the transition probability from f to any state r . There are a number of assumptions implicit in Eq. (12):

1. State f is populated only by transitions from a single initial state, i .
 2. For α_{if} to be the electron excitation rate, no other collision processes (electron capture, photo-excitation, cascade, etc.) can contribute to population of level f .
 3. No radiation field, collision process, or electric or magnetic field can modify the transition probabilities A_{fr} .
 4. Even though terms are explicitly time dependent, transport of the ions of N_i must be sufficiently slow so that those which are excited and radiate within a specific volume arrive from a region of constant plasma conditions — N_e and T_e .
- Some of these assumptions are part of what is generally called coronal equilibrium conditions. Condition 1 may be the most difficult to satisfy. Kunze discusses a case where an additional metastable level also contributes to excitation population of state f and points out that a unique solution for a given α_{if} is then not possible, though comparison with theory for some mixture is still possible. Thus the first difficulty associated with excitation rate measurements is assessing the extent to which Eq. (12) is valid.

If Expression (12) is valid for the plasma then a rate coefficient, α_{if} , can be determined by measuring the electron density, N_e ; ion density, N_i ; and absolute light intensity, I_{fj} , presuming the A_{fr} are known.

Of course the electron temperature, T_e , must also be determined in order to have α_{if} as a function of electron temperature. In practice the density N_i is inferred from the gas-filling pressure, the time evolution of light from the plasma, and a model (with some of the same assumptions about plasma conditions). That is, the N_i is determined from ionization rate measurements. N_e and T_e are generally determined by Thomson scattering of laser light. Note that I_{fj} is per unit volume requiring determination of the volume of plasma observed, which is nontrivial. If all other conditions and measurements are satisfied the absolute intensity calibration for light detection will still remain as a difficult problem with no reliable or readily transferable standard, which has been a difficulty for all types of excitation experiments. The experimental determination of excitation rate coefficients faces difficult assumptions and measurement with resulting high uncertainty, all to obtain a few data values in which much of the physically meaningful information (structure in cross section vs energy) has been averaged and lost. Nevertheless, a few excitation rate coefficients have been measured (see Refs. 1 and 20 for references to original work and some comparisons). Those rate measurements provided the first tests of excitation theory for multicharged ions.

It is currently more common to view plasma physics research as a consumer rather than a supplier of excitation cross sections. Attempts are ongoing to incorporate the best available atomic data to provide more accurate interpretation and modeling of plasmas (Refs. 13 and 22 being typical examples).

It is conceivable to use trapped ions to study electron impact excitation as has been done for ionization - Refs. 23 and 24 give reference to original work. However, many of the difficulties which apply to plasma rate measurements would apply to trapped ion measurements, and no

measurements have been reported.

3.2. Electron-Ion Beams Experiments

3.2.1. Overview of the technique

Electron-ion beams experiments were extensively developed in the 1960s and 1970s and have been reviewed.^{25,26} In addition some details are likely to be reviewed by other authors within the present volume, so that the comments given here will be restricted to an overview and a few details associated with excitation experiments.

A schematic view of a representative electron-ion crossed-beams excitation experiment is shown in Fig. 2.²⁷ The concept of the experiment is to collide electrons and ions and observe the photons of a given transition which are emitted by the decay of a particular state excited by the collision. The optical detection system consists of converging lenses, interference filter, and photomultiplier (the scintillator serves as a readily available check on relative variations in photomultiplier sensitivity). In other experiments a few different elements have been used, but typical total sensitivities of the optical system need to be about 10^{-4} counts/collision-produced photon in order for a typical experiment to be tractable.

The relationship which allows cross-section determination from experimental parameters is:

$$\sigma = \frac{S}{n} \frac{q e^2}{I_i I_e} V F \quad (13)$$

where $\frac{S}{n}$ is the rate of production of photons by the colliding beams, and the remaining terms express interacting particle density. Specifically,

I_e/e is an electron rate number from the electron current, I_e , and similarly I_i/qe for the ions; V is the relative speed of the beams in the x-y plane,

$$V = \frac{v_i v_e \sin \theta}{(v_i^2 + v_e^2 - 2v_i v_e \cos \theta)^{1/2}}$$

which reduces to just v_i for crossed beams, $\theta = 90^\circ$, with electrons much faster than ions; and the form factor, F , is the vertical (z axis) beams overlap,

$$F = \frac{\int i_e dz \int i_i dz}{\int i_e i_i n_o(z) dz}$$

where i_e and i_i are electron and ion beam vertical distributions. In Eq. (13) note that n is the probability of detecting a collision-produced photon which is dependent on where in space the photon was emitted. This vertical dependence is specifically included as $n_o(z)$ (set equal to 1 at a given height, z_0) in the form factor, and the x-y dependence has been neglected here which implies narrow crossed beams and excited-state decay times which are fast compared to motion of the excited ions. Additionally, in Eq. (13),

$$n = \left(\frac{1 - P \cos^2 \epsilon}{1 - \frac{1}{3}P} \right) D(z_0, \lambda)$$

where P is the polarization of the emitted radiation, ϵ is the direction of detection relative to polarization axis, and $D(z_0, \lambda)$ is the sensitivity of the photon detection system to isotropically emitted photons of wavelength λ originating at a central height, z_0 , within the beams collision volume.

Thus, the concept of the experiment described in Fig. 2 and

Expression (13) is quite simple but a number of detailed measurements have to be performed. The strength of the crossed-beams approach is that all of the parameters in Eq. (13) can be measured and can be independently varied in systematic tests (with sufficient patience). In practice, the absolute detection efficiency, $D(z_0, \lambda)$, is the most difficult parameter requiring elaborate measurement procedures to relate it to absolute standards that are difficult to reproduce in the laboratory with satisfactory accuracy.

3.2.2 Details of some particular cases

A primary issue in any experiment is whether or not sufficient signal, in this case photon count rate, S , will be obtained. Table 1 gives some of the parameters of two experiments, one quite tractable²⁸ (Ba^+) and the other quite difficult¹⁶ (N^{+4}). In both cases what is given is approximate "best" conditions achieved for an absolute measurement near threshold. At first glance the parameters seem quite similar, but the differences in cross-section magnitude, wavelength, and charge state of the ion modify technical difficulties significantly.

The statistical uncertainty in the primary signal, S , is given by the total accumulated counts according to

$$\Delta = \frac{\sqrt{(S + B)t}}{St}$$

where Δ is the uncertainty fraction, S and B are signal and background count rates, and t is the data collection time. Of course, in actual measurements, the signal and background must be separately accounted which generally has been accomplished by modulation of both beams and gating of counters which results in a real-time duty factor of about $\frac{1}{4}$.

Thus, actual experimental time is typically $4t$. For an arbitrary Δ of .03, $4t$ for one measurement is then 580 sec for the Ba^+ case and over 10^5 sec for the N^{+4} case. "Best" conditions are not maintained over extended times, and it is rarely satisfactory to measure one datum. Thus, for the N^{+4} case the accumulation of two absolute data points, one just below threshold (found to be zero) and the other at 15.5 eV, required about 2×10^5 sec data collection time for a $\Delta = .06$.

The N^{+4} case is useful for suggesting a few more of the difficulties associated with these experiments. Detailed systematic tests are not only possible with crossed beams, they can also be critical. Typically, cross sections are determined with deliberate variations of I_i , I_e , V , background pressure, and beams-chopping frequency because problems referred to generally as cross-beam modulation are revealed by such diagnostics. One of the beams may modify either the background or the other beam in such a way that spurious signal is produced which can either add to or subtract from the true signal. Such problems are discussed in Ref. 25 and have been encountered specifically.^{16, 29-31}

During the N^{+4} study a problem was revealed by deliberate variation of I_e . Near threshold σ was found to be independent of I_e , but at 52 eV the systematic variation of σ with I_e shown in Fig. 3 was observed. At first the problem was "guessed" to be due to redirection or focussing of the ion beam by the space charge of the electron beam. A component of ion-produced background due to ions striking surfaces had been previously identified from studies of ion noise vs background pressure, and in fact this source of background had been the cause of apparatus modifications. Thus, it was suspected that the surface noise could be modulated by electron-beam space charge redirecting ions. However, such a spurious

signal should have been independent of electron energy even to below excitation threshold since the current density of the electron beam (which would control space charge effects) changes only slightly between 10 and 50 eV energies. In fact the electron current density was deliberately changed by modifying the electron gun during the course of these studies. It was finally judged that the spurious signal was due to creation by the electron beam of some particular background target (probably an ion) which had a threshold for creation at energies above the N^{+4} excitation threshold. Light, within the bend pass of the photon system, could then be produced by this target interacting with the ion beam. At any rate the problem persisted in spite of apparatus modification, and the value of the cross section at zero electron current as extrapolated from Fig. 3 was finally accepted as the correct cross section at 52 eV.

3.3.3. Directions of beams studies

The work of Harrison and colleagues (see Ref. 25), Dolder and Peart²⁶ and, for excitation, particularly Taylor and Dunn²⁷, advanced the crossed-beams technique significantly and set high standards for detail and accuracy. The spirit of these endeavors has been to measure cross sections with the highest achievable reliability in order to provide test data against which our understanding of physical processes could be meaningfully tested and developed. Others, particularly the Russian group at Uzhgorod,³² have contributed significant data on excitation using crossed beams but have not provided sufficient detail to allow as high a level of confidence.

Experiments performed to date are nearly all similar to the description presented thus far. However, future progress may be achieved by a

few departures from these conditions. Inclined beams ($\theta \approx 8$ to 30°) have been used extensively in related types of collisions by Dolder and Peart.²⁶ Kohl and colleagues³³ have adapted inclined beams to excitation observations and attempts to study dielectronic recombination but have not yet reported cross sections. Inclined beams provide a greater beams overlap without compromising the ability to measure all of the parameters accurately.

Merged beams³⁴ are attractive from the viewpoint of beams overlap which is a primary concern. However, application of merged beams to excitation experiments will require additional technical advances. The observation of photons appears to be impractical in merged beams because of the extensive volume in which they would be produced. However, energy loss of the electrons during the collision, which has not been applied as yet in ion-excitation studies, might be coupled with merged-beams approach. Significant problems of dispersion of the electrons as a function of collision energy would have to be overcome to obtain detailed absolute cross sections from a merged-beams, energy-loss approach. However, the elimination of present problems associated with optical calibration $[D(z_0, \lambda)]$ is enticing, and the critical signal level would be increased.

3.3. Study of Excitation via Ionization

Crossed-beams studies of ionization cross sections have revealed features identified as excitation followed by autoionization. Subtracting the direct ionization cross section leaves only the excitation component. At present about as many excitation cross sections for ions can be estimated by this technique as have been measured directly.

There are a number of aspects of these excitation-autoionization measurements which are important in analyzing the results as excitation

measurements. The states excited are autoionizing resonances which are a particular class of excited states. The observed excitation-autoionization is part of a total ionization cross section and the possibility of an interference between direct ionization and the excitation component must be admitted. Additional processes such as recombination-double-autoionization³⁵ may also contribute to total ionization cross sections so that all of the increase above direct ionization may not be attributable to excitation.

In spite of these issues it is possible that meaningful tests of excitation theory can derive from excitation-autoionization measurements. A number of cases will be discussed here and some implications about excitation theory inferred based on the assumption that excitation-autoionization simply adds to ionization cross sections. Crossed-beams ionization measurements avoid problems of low photon detection efficiency and its uncertainty and high electron-produced background present in direct excitation experiments.

4. RESULTS AND DISCUSSION

4.1. Simple Systems

The simplest ion is He^+ and some of the earliest excitation experiments were for this case. The $1s - 2s$ excitation was studied experimentally by Dance et al.³¹ and by Dolder and Peart³⁶ who measured only relative cross sections due to problems of calibration at 30.4 nm, but obtained good agreement as to the shape of the cross section. However, the comparison with theory^{1,3,37-41} remains a significant point of discrepancy in the field. These results are presented in Fig. 4 which shows the more recent experiment,³⁶ normalized to close-coupling theory

at high energy, compared with a number of theoretical results. Presumably, close-coupling results should be best, but the agreement among them is poor and all the theories are higher than experiment near threshold.

The situation is better for the $1s - 2p$ excitation in He^+ in that the theories are in reasonable agreement,¹ and the experiment of Dashchenko et al.⁴² normalized to theory near 200 eV, confirms the shape predicted.

At this point it is interesting to speculate that the discrepancy in the $1s - 2s$ case can be referred back to the Hamiltonian (2) or the potential (6). The term $\frac{2}{r_{ij}}$ is expanded in a multipole representation to do the calculations of cross section. For $1s - 2p$ the dipole term dominates while for $1s - 2s$ only the monopole term contributes (except via exchange). At any rate the speculation is that the theoretical representation overestimates the monopole term. Another case will be presented later with similar implications.

The only He-like case which has been studied experimentally is Li^+ . Rogers et al.⁴³ measured the absolute emission cross section for 548.5nm which is excited by spin change (exchange), dominantly through the $1^1S \rightarrow 2^3P$ transition. The cross section exhibits structure near threshold attributed to interference involving autoionizing resonances of neutral Li (schematically illustrated in Fig. 1). The available close-coupling calculations^{1,43} do produce the observed structure. However, none of the various coupled-state and distorted-wave predictions represent the data to better than about $\pm 50\%$ over the experimental energy range 60-160 eV.

Other He-like ions have received significant theoretical attention^{1,8,44} in part because of the importance of resonances in the intercombination (spin change) and optically forbidden transitions and partly

because these ions persist over broad temperature ranges in high-temperature plasmas. Pradhan et al.⁸ predict dramatic resonance structures in these transitions so that even energy-averaged rate coefficients may be increased by factors of six or so for intercombination transitions.

The Li-like ions are perhaps the best studied experimentally and provide instructive comparison between theory and experiment. The 2s - 2p resonance lines of Be^+ (Ref. 15), C^{+3} (Ref. 45), and N^{+4} (Ref. 16) have been measured. The Be^+ results are shown in Fig. 5. None of the theories represent the experiment within experimental uncertainty (typically $\pm 10\%$ at high confidence, intended to be comparable to 98% confidence level statistics). However, with increasing power and complexity, the calculations^{46,47} approach the experiment (neglecting the $\bar{g}$ result). This case nicely demonstrates the effects of electron correlation or coupling and shows that the rigid potential screening of the Coulomb-Born approach tends to overestimate the near threshold cross section.

By contrast Fig. 6 shows results near threshold for C^{+3} . The experiment is in excellent agreement with two state close-coupling theory⁴⁸ if the experimental energy spread is folded with the theory. However, the Coulomb-Born⁴⁸ with exchange calculation is just as acceptable within the experimental uncertainty. For N^{+4} (Fig. 3) the situation is nearly identical to C^{+3} . The electronic structure is constant for these cases, but the relative spacing of energy levels is significantly different (see Fig. 7). This energy scaling alone probably does not account for the dramatic decrease in effects of coupling along the sequence. The electron correlation part of the potential [integral part of Eq. (6)] is generally weaker compared to the nuclear part as ionic charge increases

along the isoelectronic sequence, and this is probably the dominant effect. In order to assess the extent to which distorted-wave or Coulomb-Born approximations are valid for highly charged ions, it remains important to measure cross sections along isoelectronic sequences other than Li-like where the scaling of energy levels will not correspond to Fig. 7.

The experimental study of ionization of Li-like ions^{49,50} has produced information on excitation of the form $1s^2 2s \rightarrow 1s 2s 2\ell$ where $\ell = s$ or p . Figure 8 shows the N^{+4} case in detail, and Table 2 gives experimental cross sections deduced from simply subtracting the excitation-autoionization bumps from an extrapolated smooth direct ionization curve near the excitation threshold and comparing the experimental and theoretical results.^{51,52} The Be^+ , C^{+3} , and N^{+4} results compare reasonably with close-coupling theory, but the O^{+5} case presents a significant discrepancy. Uncertainties of these experimental excitation cross sections are difficult to assess because there are both assumptions and subtractions in the determination. However, the magnitude of inner-shell excitation, the interaction of excitation and ionization, and the experiment for O^{+5} are open to question.

4.2. Complex Systems

Some of the best experimental measurements have been for heavy ions for which the wavelength and cross-section magnitude simplify some of the technical difficulties. The excitation of the $4s - 4p$ resonance lines of Ca^+ by Taylor and Dunn²⁷ must rank as one of the most detailed experiments to be accomplished in atomic collisions studies. Crossed-beams experiments for Mg^+ , Ca^+ , Sr^+ (Ref. 32), Ar^+ , and Kr^+ (Ref. 55), Hg^+ (Refs. 56 and 57), and Zn^+ (Ref. 58) have been reported. The $6s - 6p$

resonance transitions of Ba^+ have been studied by the greatest number of experimenters.^{28,32,59-61}

For the present discussion Ba^+ will be taken as a representative case. Figure 9 shows the energy level distribution and the $6^2\text{S}_{1/2} \rightarrow 6^2\text{P}_{3/2}$ excitation cross section derived from the 455.4nm emission cross section after corrections for cascade, metastable ions in the beam, and branching ratio of the decay.²⁸ The effect of resonances is clear in the experimental cross section and is even more obvious in the polarization of the radiation. The coupled-channel technique is probably appropriate for this case but has not been attempted because of the complexity. The result that distorted wave theory⁶² overestimates excitation for complex, singly-charged ions appears to be general for all the cases cited.

The magnitude of excitation-autoionization increases for the alkali-like ions with increasing numbers of electrons^{26,63} as shown in Fig. 10. The excitation-autoionization is substantial for Ba^+ , and the excitations of the form $5p^66s \rightarrow 5p^55d6s$ can be estimated from the data. The first attempt at calculating the excitation cross sections⁶⁴ did not account for the structure within the configurations and neglected some of the important dynamic effects (exchange and nondipole transitions). The result was artificially good agreement of theory and the experiment for the excitation component. Hansen⁶⁵ demonstrated that the original analysis was in error due to neglect of structure so that the transition for which the cross section was calculated⁶⁴ would not occur at the energy of excitation-autoionization onset in the experiment. Ongoing structure calculations and distorted wave prediction of cross sections⁶⁶ indicate that excitation of a number of terms contributes and that exchange and other complexities must be included to reasonably represent the total

excitation cross section for $5p^6 6s \rightarrow 5p^6 5d 6s$ which can then be more meaningfully compared with experiment.

Recent work on ionization of alkali-like ions Ti^{+3} , Zr^{+3} , and Hf^{+3} has observed that near threshold the ionization cross sections are almost entirely due to excitation-autoionization.⁶⁷ Figure 11 shows that for Ti^{+3} the excitation-autoionization is about ten times the direct cross sections. Distorted wave calculations of the excitations of $3p^6 3d \rightarrow 3p^5 3d^2$ have been divided by 2.5 to provide comparison with experiment and are also convoluted with a 2-eV energy spread representative of the experiment. In spite of the significant discrepancy in magnitude, the energy location and relative contributions of different levels are in such good agreement as to provide promise that theory can be developed to predict such excitation cross sections for complex ions.

Similar work on excitation-autoionization for Ga^+ (Ref. 68) is also providing insight into excitation via excitation-autoionization. Further, experiments on Zn^+ for both direct excitation of bound states⁵⁸ and excitation-autoionization⁶⁸ provides an additional test case where accurate experimental data are available.

Work on excitation-autoionization of Na-like ions⁶⁹⁻⁷² has provided data on excitation of the configurations $2p^6 3s \rightarrow 2p^5 3s 3p$. This system is somewhat simpler than the more complex alkali-like cases, but the excitations are $\Delta n = 1$ transitions so that the relative intensity is smaller. Figure 12 compares a recent experiment⁷¹ and distorted wave theory⁷² for Al^{+2} . It is apparent that the $2p^6 3s \rightarrow 2p^5 3s 3p$ transition, which is predicted to be the largest component, is missing or quite small in the experiment. The same result is even more obvious in the Mg^+ case⁶⁹⁻⁷² and occurs in both distorted wave⁷² and Coulomb-Born⁷⁰

approximations. An interesting perspective is that the strength of the 2p - 3p transition derives from the monopole term in the multipole expansion of the potential (6) which is similar to the 1s - 2s excitation of He^+ which remains in discrepancy.

5. CONCLUSIONS AND DIRECTIONS

Most of the experimental data on electron impact excitation of bound states of ions has been for resonance lines of ions with one electron outside a closed shell. Nevertheless, these experiments are mostly for complex or many-electron cases, while initial detailed theoretical work was for simple, few-electron systems, so that comparisons producing physical insight have been slow to develop. Theory has attained a sufficient level of maturity and can undertake more complex systems, while recent experiments, such as those on Li-like and He-like ions, have provided more direct experimental data for detailed comparison with theory.

However, excitation experiments are to some extent stalled at present against technical difficulties of low signal and difficult calibration. Experimental progress depends on development of new approaches including new means of observing bound states as well as observations like excitation-autoionization resonances. The possibility of interference between direct ionization and excitation-autoionization complicates the comparisons and at the same time presents a new physical problem. Theoretical techniques to study interferences in the ionization channels are in development.⁷³

The interactive nature of the processes of excitation, recombination, and ionization have become more obvious in theory, experiment, and applications of the basic data. These interactions are most apparent

for multi-charged ions where the transition probabilities of these different processes become comparable. The current theoretical study of LaGattuta and Hahn³⁵ illustrates this issue. Figure 13 shows their present prediction of the electron impact ionization of Fe^{+15} which assumes simple addition of direct ionization, excitation-autoionization, and dielectronic-recombination-double-autoionization. The last process had not previously been proposed but arises through the resonances below each inner-shell excitation which are doubly excited states of the recombined ion Fe^{+14} . A dominant mode of relaxation of these highly excited resonances is predicted to be double autoionization so that dielectronic recombination now makes an appearance in the ionization cross section.

It seems clear that a significant amount of interesting physics is not yet well understood in collisions of electrons and ions. However, it is a difficult challenge to experimental techniques to provide any data, especially for excitation, which can test understanding.

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At the completion of ~~the~~ this manuscript it was discovered that a similar but broader review by G. H. Dunn,⁷⁴ had previously been prepared

for the conference on "Physics of Ionized Gases, 1980" held in Yugoslavia. Publication of those proceedings is forthcoming and Dunn's paper should be interesting to anyone following the field.

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TABLE 1. Parameters from two electron impact excitation experiments.

Parameter	Ba ⁺ (Ref. 28)	N ⁴⁺ (Ref. 16)
Electron Energy	4 eV	15.5 eV
Electron Current	10 μ A	90 μ A
Ion Energy	750 eV	40 keV
Ion Current	0.1 μ A	1.0 μ A
Pressure (both beams on)	1×10^{-9} torr	1.2×10^{-9} torr
Wavelength	455 nm	124 nm
Band Pass	10 nm	30 nm
$D(z_0, \lambda)$	7.4×10^{-4}	4×10^{-4}
Signal (S) (both beams)	10 Hz	2 Hz
Background (B) (both beams)	3 Hz	90 Hz
Emission Cross Section	$17.4 \times 10^{-16} \text{ cm}^2$	$2.7 \times 10^{-16} \text{ cm}^2$

TABLE II. Comparison of theoretical and experimental cross sections for inner-shell excitation of Li-like ions, $1s^2 2s \rightarrow 1s2s2l$ at energies about 1.1 times excitation threshold.

Ion	Energy (eV)	Cross Sections in 10^{-19} cm^2			
		Experiment ^a	Scaled Coulomb-Born ^b	Six-state close-coupling ^c	Ratio Exp/6cc Theory
Be ⁺	130	17	23	12.2	1.4
C ⁺³	340	3.2	3.7	2.15	1.5
N ⁺⁴	460	1.8	2.0	1.27	1.4
O ⁺⁵	612	2.8 (1.4)	1.1	0.74	3.8 (1.9)

- ^a. Cross sections determined by estimating the increase in total ionization cross section at energy 1.1 times excitation threshold. The Be⁺ case is from Ref. 50 while C⁺³, N⁺⁴, and O⁺⁵ are from Ref. 49. For O⁺⁵ the uncertainties are large and the value in parenthesis is the smallest excitation cross section derived from the ionization data allowing for the 90% confidence level error bars.
- ^b. From Ref. 52 which cautions that the technique is not appropriate to low ionic charge cases such as Be⁺.
- ^c. From Ref. 51 with Be⁺ case extrapolated according to $(z - 1.4)^3 \sigma = \text{constant}$ for a given energy in threshold units.

FIGURE CAPTIONS

Fig. 1. Schematic representation of the energy levels of the electron + He-like ion. The state of the He-like ions is designated with k representing the third electron which is either in bound states or the continuum. The arrow indicates an energy for which continuum plus ground $1S$ (elastic scattering), continuum plus $3S$ (excitation), or bound plus $1S$ (resonance) are all possible states of the system.

Fig. 2. Schematic of a representative electron-ion crossed-beams excitation experiment (from Ref. 27).

Fig. 3. a.) Cross section for electron impact excitation of $N^{+4}(2s - 2p)$ from Ref. 16. The solid point at 15.5 eV is the absolute measurement with outer error bar showing total absolute uncertainty at 90% confidence level - other points have 90% confidence level relative bars. The cross sections at 26 and 52 eV have been corrected for variation with electron current. The dashed curve is coupled-state theory by Van Wyngaarden and Henry (see Ref. 1).

b.) Apparent cross section at 52 eV as a function of electron current. For the solid points the electron current density was increased about 50% by decrease of electron beam height. Error bars here are 1 std. dev. statistics.

Fig. 4. Collision strength for electron impact excitation of $He^+(1s - 2s)$ as a function of threshold energy ϵ from Ref. 1. Solid points connected by dashed line are experiment (Ref. 36) normalized to theory B at high energy. Theoretical results are: A - 3 state close-coupling (3cc) of Ref. 37; B - 3cc of Ref. 3; C - distorted wave of Ref. 38; D - distorted wave second-order potential of Ref. 39; E - 5cc of Ref. 40; and F - 3cc of Ref. 41.

Fig. 5. Electron impact excitation of $\text{Be}^+(2s - 2p)$ - from Ref. 15. Experiment is dashed curve with typical high confidence uncertainty indicated. Theories from Refs. 46 and 47 are non-unitarized Coulomb-Born (CBI with CBXI including exchange); unitarized Coulomb-Born (CBII and CBXII); unitarized distorted wave (UDWPOII); and close-coupling (2cc and 5cc). The $\bar{g}$ approximation is according to Ref. 11.

Fig. 6. Electron impact excitation of $\text{C}^{+3}(2s - 2p)$ near threshold from Ref. 45. Theoretical results are from Ref. 48. The solid curve through the data is the 2 state close-coupling convoluted with the experimental energy spread. Error bars are 90% confidence level statistics except for the outer bar at 10.2 eV which is total absolute uncertainty including absolute optical calibration at 90% confidence level.

Fig. 7. Energy levels of the Li-like ions scaled to the energy of the $2s - 2p$ resonance transition, from Ref. 16.

Fig. 8. Electron impact ionization of N^{+4} showing contribution due to $1s^2 2s \rightarrow 1s 2s n l$ inner-shell excitation-autoionization - from Ref. 49. The excitation calculation of 6cc (Ref. 51) for $1s^2 2s \rightarrow 1s 2s 2 l$ only is represented by the solid curve beginning at 415 eV and has been added to the dashed curve which is scaled-Coulomb-Born direct ionization calculation of Ref. 54. Solid curve below 350 eV is Coulomb-Born direct ionization of Ref. 51.

Fig. 9. Electron impact excitation of Ba^+ for $6^2S_{1/2} \rightarrow 6^2P_{3/2}$ and energy levels of Ba^+ with the transitions studied experimentally indicated by boxes - from Ref. 28. Solid curve is experiment of Ref. 28 with relative (inner) and absolute (outer) error bars at 98% confidence level. Open circles (Ref. 59) and open triangles (Ref. 60) are earlier measurements. Distorted wave theory (Ref. 62) is indicated by solid points.

Fig. 10. Electron impact ionization of alkali-like ions showing significant enhancement due to excitation-autoionization at abrupt rises near 15, 23, and 27 eV for Ba^+ , Sr^+ , and Ca^+ respectively - from Ref. 63.

Fig. 11. Electron impact ionization of Ti^{+3} from Ref. 67. Dashed curve is the Lotz formula prediction taken to be the direct ionization. The solid curve is a distorted wave-dipole only calculation divided by 2.5 and added to the Lotz prediction. Dot-dashed curve is the solid curve convoluted with 2 eV energy spread for comparison to the experiment.

Fig. 12. Electron impact ionization of Al^{+2} showing distinct contribution due to excitation-autoionization - from Ref. 71. Solid curve is distorted wave ionization theory (normalized to the experiment at 70 eV) with distorted wave excitation of the form $2p^6 3s - 2p^5 3sn\hat{n}$ (Ref. 72) added to the ionization theory.

Fig. 13. Electron impact ionization of Fe^{+15} from Ref. 35. Solid curve is Lotz formula for direct ionization with dashed curve added to represent excitation-autoionization contribution and then the hatched area added to represent dielectronic recombination-double autoionization contribution.

Figure 1.
5

$$\Delta E(2s - nI) / \Delta E(2s - 2p)$$

ENERGY LEVELS OF Ba⁺

Fig. 1