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Population Distributions in Charge Exchange. I. Proton Capture Cross Sections for Levels $n=1$ Through $n=15$

JOHN R. HISKES*
Culham Laboratory, United Kingdom Atomic Energy Authority, Abingdon, Berks, England

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The charge-exchange capture cross sections are calculated for captures into all states of the hydrogen atom for levels through $n=15$ using the Brinkman-Kramers matrix element. These expressions are sufficiently general to simulate captures from all the elements, assuming only one electron is active.

1. INTRODUCTION

ELECTRON capture by protons passing through gases is known to result in captures into all states of the atom. Early calculations[1,2] had shown that, at high-proton velocities, captures into s states predominate, and had indicated a population distribution which varied inversely with the third power of the principal quantum number. Later work[3,4] has shown that in general the population distributions will be functions of five parameters involved in the capture process: the effective charges of both the target and the incident projectile, the principal quantum number and angular momentum of the active electron, and the relative velocity of the incident projectile and target. Although expressions for electron-capture cross sections through principal quantum level $n=4$ have been available,[4] there has been little explicit discussion in the literature of the dependence of the population distribution on the above-mentioned parameters.

For the most part, discussion of excited-state capture has been based on the Born matrix element, and the Brinkman-Kramers (BK) approximation to the Born expressions.[3–5] While these approximations are known to give poor results for absolute values of the capture cross sections, there is some evidence to suggest that the BK matrix element will prove reliable for ratios of the capture cross sections. In recent experiments with thin hydrogen targets, Riviere and Sweetman[6,7] have found the population distributions follow closely an n^{-3} distribution for levels $n=9$ through $n=23$, and agree with the BK ratio for captures into the $n=14$ level for energies above 40 keV.

An interest in the population distributions for hydrogen has arisen recently in conjunction with experiments designed to produce thermonuclear plasmas. Beams of excited energetic hydrogen atoms are expected to play an important role in contemporary neutral injection experiments.[8–10] In these injection experiments the excited levels below $n=6$ are not expected to contribute significantly because of their relatively short radiative lifetime,[11] while those levels above about $n=15$ are generally not of interest because of their rather low electric ionization thresholds.[12] It is this group of approximately ten levels, from $n=6$ through $n=15$, which, at the present time, appears to be of primary interest for neutral injection.

Earlier, an impact-parameter method[13] was used to calculate the s-state capture cross section for the ten lowest levels of hydrogen in the case of a lithium or cesium neutralizer.[14] The use of lithium vapor as a neutralizer had been proposed previously as a means for enhancing the population of the $H(2S)$ state.[15] An experiment by Futch and Damm[16] using protons incident on lithium vapor has shown that excited-state populations for levels $n=8$, 9, and 10 exhibit an increase similar to that indicated by the theory. Since the impact parameter method is based on physical assumptions similar to those leading to the BK matrix element, their experiment suggests that for purposes of comparing the population distributions for different neutralizers the BK approximation may be useful for energies as low as 20 keV. A difficulty with the impact-parameter method as applied to charge exchange is that in general it does not as readily lead to closed expressions for the cross sections as does the BK method. Only in

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* Present address: Lawrence Radiation Laboratory, Livermore, California.

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the limit of high energies, corresponding to small impact parameters, can the integrals be easily evaluated without recourse to numerical methods.[17] For this reason, the BK matrix element would appear to be more suited as a basis for a theoretical survey of neutralizers than would the impact-parameter method.

In this paper is discussed the capture cross sections for all states through the $n=15$ level in the case of protons incident on various atomic gases. Capture cross sections are presented which are derived using the BK matrix element and for a range of effective nuclear charge Z, target principal quantum level n, and angular momentum l, sufficient to simulate all the elements in a one electron approximation. It is believed these cross sections will aid in elucidating those regions of the periodic table where significant population enhancement might occur, and will give some indication of the magnitude of the population enhancement. In previous papers dealing with excited-state calculations the relevant integrals have been evaluated by means of parametric differentiation.[4,5,18] The extension of these calculations to the very highly excited levels using this method would involve considerable labor. Here we utilize the fact that the integrals can be expressed in terms of momentum eigenfunctions which in turn are readily generated using recursion relations.

2. QUANTITATIVE DISCUSSION

In this section is considered the electron-capture cross sections for protons incident on an atomic neutralizer in which it is assumed that only a single outer electron is active. The BK expression of the cross section for capture into the state n,l of the hydrogen atom from a state ν, λ of a neutralizer in which the electron is moving initially in a Coulomb potential with effective charge eZ, is given by[4]

$$\sigma(\nu\lambda-nl)=(2\pi)^{-1}M^2\hbar^{-4}(v_f/v_i)\int_{-1}^{+1}|\mathfrak{M}|^2d(\cos\theta)\,,\qquad(1)$$

where

$$|\mathfrak{M}|^2=\left|\int\phi(\nu\lambda)(e^2Z/r)\exp i\bar{\alpha}\cdot\bar{r}d\bar{r}\right|^2\times\left|\int\phi^*(nl)\exp i\bar{\beta}\cdot\bar{s}d\bar{s}\right|^2.\qquad(2)$$

The angle θ is the angle between the initial and final velocities $\bar{v}_i$ and $\bar{v}_f$, and M is the reduced mass. The other quantities are defined by

$$\bar{\alpha}=\bar{k}_f+\bar{k}_iM_1/(M_1+m)\,;$$
$$\bar{\beta}=-\bar{k}_i-\bar{k}_fM_2/(M_2+m)\,;$$
$$\bar{k}_i=\bar{v}_i\hbar^{-1}(M_1+m)M_2/(M_1+M_2+m)\,;$$
$$\bar{k}_f=\bar{v}_f\hbar^{-1}(M_2+m)M_1/(M_1+M_2+m)\,.$$

The masses M_1, M_2, and m are the target nuclei, proton, and electron masses, respectively.

Introducing a new integration variable

$$y=\alpha^2a_0^2+a^2=\beta^2a_0^2+b^2\,,$$

where $a=Z/\nu$, $b=1/n$, and a_0 the Bohr radius, Eq. (1) can be written

$$\sigma(\nu\lambda-nl)=(4\pi a_0^2p^2e^4)^{-1}\int_d^\infty|\mathfrak{M}|^2dy\,;\qquad(3)$$

here the upper limit is taken to be infinite as a matter of convenience. The quantities p^2 and x are defined by

$$p^2=(mv_ia_0\hbar^{-1})^2\,,$$
$$x=\tfrac{1}{4}p^2+\tfrac{1}{2}(a^2+b^2)+(b^2-a^2)^2/4p^2\,.$$

The momentum eigenfunctions are defined as[19]

$$\Phi(nlm)=(2\pi)^{-3/2}\int\phi(nlm)e^{-i\bar{k}\cdot\bar{r}}d\bar{r}\,.\qquad(4)$$

For a Coulomb potential these functions can be written

$$\Phi(nlm)=F(nl)Y(lm)\,,\qquad(5)$$

where the $Y(lm)$'s are the spherical harmonic and the general form for the function $F(nl)$ is given by Bethe and Salpeter.[19]

Writing Eq. (2) as

$$|\mathfrak{M}|^2=|\psi(\nu\lambda)|^2|\chi^*(nl)|^2$$

and comparing Eqs. (2), (4), and (5), we see that

$$|\chi^*(nl)|^2=2\pi^2(2l+1)a_0^3F^2(nl)\,.\qquad(6)$$

Using[20]

$$|\psi(\nu\lambda)|^2=(4a_0^2)^{-1}e^4y^2|\chi(\nu\lambda)|^2\,,\qquad(7)$$

the expression (3) for the capture cross section can be written

$$\sigma(\nu\lambda-nl)=\pi^3a_0^2(2l+1)(4p^2Z^3)^{-1}\times\int_x^\infty y^2F^2(\nu\lambda)F^2(nl)dy\,.\qquad(8)$$

Expressions for the cross sections have been obtained using Eq. (8) for all n,l states through the $n=15$ level and for ν,λ states $1s\rightarrow 7s$, $2p\rightarrow 6p$, and $3d\rightarrow 5d$, inclusive. Following Bates and Dalgarno (BD),[4] advantage is taken of the fact that considerable compactness can be achieved by expressing these cross sections as

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simple integrals. Writing Eq. (8) in the form

$$\sigma(\nu\lambda-nl)=\pi a_0^2 p^{-2} D(\nu\lambda-nl)\int_x^{\infty} G(\nu\lambda-nl)dy, \quad (9)$$

the appropriate quantities are

$$D(1s-ns)=2^8Z^5n^{-5}.$$

On writing $z=y^{-1}(y-2b^2)$ and defining the Gegenbauer polynomial, C_{n-l+1}^{l+1}, (Ref. 19, p. 39), the expression for G becomes

$$G(1s-1s)=y^{-6}C_{n-1}^{1}.$$

Continuing,

$$D(1s-np)=(3)2^{12}Z^5n^{-7}(n^2-1)^{-1};$$
$$G(1s-2p)=y^{-8}(y-b^2)C_{n-2}^{2}.$$

$$D(1s-nd)=(5)2^{18}Z^5n^{-9}(n^2-4)^{-1}(n^2-1)^{-1};$$
$$G(1s-3d)=y^{-10}(y-b^2)^2C_{n-3}^{3}.$$

$$D(1s-nf)=(7)2^{22}3^2Z^5n^{-11}(n^2-9)^{-1}(n^2-4)^{-1}(n^2-1)^{-1};$$
$$G(1s-4f)=y^{-12}(y-b^2)^3C_{n-4}^{4}.$$

$$D(1s-ng)=2^{30}3^4Z^5n^{-12}[(n-5)!][(n+4)!]^{-1};$$
$$G(1s-5g)=y^{-14}(y-b^2)^4C_{n-5}^{5}.$$

$$D(1s-nh)=(11)2^{34}3^25^2Z^5n^{-14}[(n-6)!][(n+5)!]^{-1};$$
$$G(1s-6h)=y^{-16}(y-b^2)^5C_{n-6}^{6}.$$

$$D(1s-ni)=(13)2^{40}3^45^2Z^5n^{-16}[(n-7)!][(n+6)!]^{-1};$$
$$G(1s-7i)=y^{-18}(y-b^2)^6C_{n-7}^{7}.$$

$$D(1s-nk)=(15)(7!)^22^{36}Z^6n^{-18}[(n-8)!][(n+7)!]^{-1};$$
$$G(1s-8k)=y^{-20}(y-b^2)^7C_{n-8}^{8}.$$

$$D(1s-nl)=(17)(8!)^22^{40}Z^5n^{-20}[(n-9)!][(n+8)!]^{-1};$$
$$G(1s-9l)=y^{-22}(y-b^2)^8C_{n-9}^{9}.$$

$$D(1s-nm)=(19)(9!)^22^{44}Z^5n^{-22}[(n-10)!][(n+9)!]^{-1};$$
$$G(1s-10n)=y^{-24}(y-b^2)^9C_{n-10}^{10}.$$

$$D(1s-nn)=(21)(10!)^22^{48}Z^5n^{-24}[(n-11)!][(n+10)!]^{-1};$$
$$G(1s-11n)=y^{-26}(y-b^2)^{10}C_{n-11}^{11}.$$

$$D(1s-no)=(23)(11!)^22^{52}Z^5n^{-26}[(n-12)!][(n+11)!]^{-1};$$
$$G(1s-12o)=y^{-28}(y-b^2)^{11}C_{n-12}^{12}.$$

$$D(1s-nq)=(12!)^22^{56}5^2Z^5n^{-28}[(n-13)!][(n+12)!]^{-1};$$
$$G(1s-13q)=y^{-30}(y-b^2)^{12}C_{n-13}^{13}.$$

$$D(1s-nr)=(13!)^22^{60}3^3Z^5n^{-30}[(n-14)!][(n+13)!]^{-1};$$
$$G(1s-14r)=y^{-32}(y-b^2)^{13}C_{n-14}^{14}.$$

$$D(1s-nt)=(29)(14!)^22^{64}Z^5n^{-32}[(n-15)!][(n+14)!]^{-1};$$
$$G(1s-15t)=y^{-34}(y-b^2)^{14}C_{n-15}^{15}.$$

Defining $w=y^{-1}(y-2a^2)$, the capture cross section for those neutralizers in which the active electron is not initially in the $1s$ state can be derived from

$$D(2s-nl)=2^{-3}D(1s-nl);$$
$$G(2s-nl)=y^{-2}(y-2a^2)^2G(1s-nl).$$

$$D(3s-nl)=3^{-5}D(1s-nl);$$
$$G(3s-nl)=(4w^2-1)^2G(1s-nl).$$

$$D(4s-nl)=4^{-5}D(1s-nl);$$
$$G(4s-nl)=(8w^3-4w)^2G(1s-nl).$$

$$D(5s-nl)=5^{-5}D(1s-nl);$$
$$G(5s-nl)=(16w^4-12w^2+1)^2G(1s-nl).$$

$$D(6s-nl)=6^{-5}D(1s-nl);$$
$$G(6s-nl)=(32w^5-32w^3+6w)^2G(1s-nl).$$

$$D(7s-nl)=7^{-5}D(1s-nl);$$
$$G(7s-nl)=(64w^6-80w^4+24w^2-1)^2G(1s-nl).$$

$$D(2p-nl)=2^{-3}3^{-1}Z^2D(1s-nl);$$
$$G(2p-nl)=y^{-2}(y-a^2)G(1s-nl).$$

$$D(3p-nl)=(2)3^{-7}Z^2D(1s-nl);$$
$$G(3p-nl)=y^{-2}(y-a^2)(4w)^2G(1s-nl).$$

$$D(4p-nl)=2^{-10}3^{-1}5^{-1}Z^2D(1s-nl);$$
$$G(4p-nl)=y^{-2}(y-a^2)(12w^2-2)^2G(1s-nl).$$

$$D(5p-nl)=(2)3^{-1},5^{-7}Z^2D(1s-nl);$$
$$G(5p-nl)=y^{-2}(y-a^2)(32w^3-12w)^2G(1s-nl).$$

$$D(6p-nl)=2^45^{-1}6^{-7}7^{-1}Z^2D(1s-nl);$$
$$G(6p-nl)=y^{-2}(y-a^2)(80w^4-48w^2+3)^2G(1s-nl).$$

$$D(3d-nl)=2^73^{-9}5^{-1}Z^4D(1s-nl);$$
$$G(3d-nl)=y^{-4}(y-a^2)^2G(1s-nl).$$

$$D(4d-nl)=2^{-10}3^{-2}5^{-1}Z^4D(1s-nl);$$
$$G(4d-nl)=y^{-4}(y-a^2)^2(6w)^2G(1s-nl).$$

$$D(5d-nl)=2^73^{-2}5^{-9}7^{-1}Z^4D(1s-nl);$$
$$G(5d-nl)=y^{-4}(y-a^2)^2(24w^2-3)^2G(1s-nl).$$

Explicit expressions for these cross sections have been given elsewhere.[21] In labeling these states the spectroscopic notation given in Condon and Shortley[22] has been used. It is easily verified that the above expressions reduce to those given by BD for states through $n=4$.

In a subsequent paper explicit results of the population distributions for a variety of neutralizers will be presented. Here we restrict the discussion to a few semiquantitative remarks based on the general form of

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the capture cross sections as given by Eq. (8). First, for a sufficiently high incident-proton velocity and for all neutralizers we have the well-known result that the s-state capture distribution varies as n^{-3}, as deduced from the coefficient of the asymptotic form for $F^2(nl)$. Second, captures into the very high angular-momentum states are in general not expected to contribute appreciably to the total cross section since the magnitude of $F^2(nl)$ is dominated by the coefficient

$$2^{4l}(l!)^2[(n-l-1)!]^2[(n+l)!]^{-2}.$$

In this connection, we note that the large resonance reported by Butler and Johnston[23] at $p^2=1$ is not reproduced in these calculations.

Note added in proof: This resonance has been discussed further by S. T. Butler, R. M. May, and I. D. S. Johnston, Phys. Letters **10**, 281 (1964).

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Generalization of the "Linear Theory" of Configuration Interaction*

B. G. Wybourne
Chemistry Division, Argonne National Laboratory, Argonne, Illinois

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The "linear" theory of configuration interaction has met with considerable success in giving a phenomenological description of the energy levels of equivalent-electron systems. In the present paper the generalization of the "linear" theory to configurations containing nonequivalent electrons is examined from the point of view of second-order perturbation theory. It is shown that most second-order electrostatic interactions can be phenomenologically described by the first-order terms of a set of effective two-body scalar interactions. The significance of these interactions in atomic energy-level calculations is considered.

I. INTRODUCTION

It is well known[1] that the solutions of the Hartree-Fock equations for complex atoms or ions yield multiplet energy separations that are considerably *larger* than those found experimentally. The discrepancies are usually so great that these calculations are of little value in the prediction and correlation of atomic energy levels. As a result, atomic spectroscopists have tended to correlate their observations with theoretical energy levels calculated by constructing the energy matrices for the relevant electron configurations and then treating the radial integrals as phenomenological parameters.[2] The parameters are usually found to be substantially *smaller* than the Hartree-Fock radial integrals. The agreement between the theoretical and experimental energy levels has frequently been strikingly close considering that in most cases configuration interaction has been explicitly ignored. It is as if the parameters have adjusted themselves so as to accommodate part of the effects of configuration interaction.

The "linear" theory of configuration interaction has been a natural outgrowth of an early paper of Bacher and Goudsmit[3] who demonstrated that most, though not all, of the second-order electrostatic interactions can be added linearly. Originally Bacher and Goudsmit used linear relations to express the unknown energy levels in terms of the *observed* energy levels of the atom and its ions. Later developments by Trees[4–9] and Racah[10–13] have sought to replace the second-order effects by the first-order terms of an effective two-body interaction. A detailed analysis of the physical content of these interactions in l^N-type configurations has been given by Rajnak and Wybourne.[14–17] The use of effective two-body interactions has found extensive application

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