Electron Capture into High Excited States in p + H Collisions*

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Differential and integrated cross sections for proton–hydrogen-atom charge-exchange collisions are calculated for capture into highly excited states using a method which takes the nonorthogonality of initial and final states into account. The differential cross sections show dramatic features that persist from charge exchange into the lower quantum states. The integrated-cross-section results agree well with experiments over the range of validity of the method (incident proton energies above 20 keV) and are a considerable improvement over previous calculations, which are an order of magnitude larger than experiment. The postulated scaling rules of Jackson and Schiff and of Omidvar are compared with the calculations, and the former is in better agreement.

In this paper we report the results of calculations on the production of highly excited states of hydrogen atoms by the electron-capture process

$$p + \mathrm{H}(1s) \rightarrow \mathrm{H}(nlm) + p\,. \tag{1}$$

The calculations are based on a recently developed method which we have described elsewhere[1,2] for calculating rearrangement collisions.

Khayrallah, Karn, Koch, and Bayfield[3] observed the production of highly excited states resulting from reaction (1). They observed the cross section for electron capture into all states with quantum numbers $13 \leq n \leq 30$. The quantity $\sigma_*(E)$ is defined as the sum of the cross sections into all these states at the incident proton energy E. Khayrallah *et al.* obtained $\sigma_*(E)$ in the energy range $10 \leq E \leq 60$ keV. When $\sigma_*(E)$ is calculated by the method of Butler, May, and Johnston,[4] there is an order-of-magnitude discrepancy between theory and experiment. Our own computed results are entirely compatible with the results of Khayrallah *et al.* over the expected range of validity of our method. The predictions of Omidvar,[5] that the electron-capture cross sections scale as $1/n^2$, as opposed to the $1/n^3$ scaling postulated by Jackson and Schiff,[6] will also be discussed.

Based on the method of I and II, the transition matrix element for electron-capture collisions at a proton energy E is given by

$$T_{nlm}(E,\lambda,\Phi) = \left(a(n,C^2)\varphi^*_{nlm}(\vec{\mathrm{C}})\varphi_{100}(\vec{\mathrm{B}}) + \frac{1}{2\pi^2}\int \frac{d^3k}{k^2}\varphi^*_{nlm}(\vec{\mathrm{C}}-\vec{\mathrm{k}})\varphi_{100}(\vec{\mathrm{B}}-\vec{\mathrm{k}})\right)\Big/(2\pi)^3[1-|\varphi^*_{nlm}(\vec{\mathrm{C}})\varphi_{100}(\vec{\mathrm{B}})(2\pi)^{-3}|^2]\,, \tag{2}$$

where $\varphi_{nlm}(\vec{\mathrm{p}})$ are hydrogenlike wave functions in momentum space, $\vec{\mathrm{B}}$ and $\vec{\mathrm{C}}$ are the linear combinations of initial and final momenta $\vec{\mathrm{k}}_i$, $\vec{\mathrm{k}}_f$,[7]

$$\vec{\mathrm{B}} = \vec{\mathrm{k}}_f - \vec{\mathrm{k}}_i[1 - 1/(M_p+1)]\,, \tag{3}$$

$$\vec{\mathrm{C}} = \vec{\mathrm{k}}_f[1 - 1/(M_p+1)] - \vec{\mathrm{k}}_i\,, \tag{4}$$

λ is related to the C.M. scattering angle θ by the equation

$$\lambda = 4(M_p+1)^2 \sin^2\tfrac{1}{2}\theta\,, \tag{5}$$

Φ is the azimuthal scattering angle, and

$$a(n,C^2) = -\tfrac{1}{2}(C^2 + 1/n^2 + 1/2\pi^2)\,. \tag{6}$$

For explicit expressions see Eq. (33)–(39) of I. Included in the formula for T_{nlm} are contributions needed to take into account the non-orthogonality of initial and final states of the electron. These terms are the factor in square brackets in the denominator and the last term in function $a(n,C^2)$. They have been important in bringing calculated cross sections into agreement with experiments on the total and the low-excited-state charge-transfer reactions[1,2] and are of particular importance in the region of incident energy between 25 and 100 keV. The term in T_{nlm} given by the integral over $\vec{\mathrm{k}}$ arises from the proton-proton interaction potential energy.

With the calculational method proposed in II we are able to compute the transitional matrix element, even for capture into very highly excited states. Previous attempts at calculating the transition matrix element relied upon the Feynman auxiliary integral technique[8,9] which becomes unmanageable for the highly excited states, especially when the non-orthogonality correction terms are included in the calculation. The present method consists of rotating the coordinate system in such a way that the vectors $\vec{\mathrm{B}}$ and $\vec{\mathrm{C}}$ lie in the x-z plane and $\vec{\mathrm{C}}$ lies along the $-\hat{z}$ axis, since in this

coordinate system the term in the transition matrix element arising from the proton-proton interaction is easier to handle. For details of the method, see Sec. 2 of II.

The conditions for the validity of Eq. (1) are considered at length in I. For the present electron-capture process the conditions are satisfied for incident proton energies above 25 keV. We calculate the differential cross section for capture into nlm states summed over all m,

$$\frac{d\sigma_{nl}(E)}{d\Omega} = (2\pi)^4 \frac{k_f}{k_i}\left(\frac{M_p}{2}\right)^2 \times \sum_{m=-l}^{l} |T_{nlm}(E,\lambda,\Phi)|^2 , \tag{7}$$

and integrate over all solid angles to obtain the integrated charge exchange cross section

$$\sigma_{nl}(E) = \frac{2\pi}{2M_p^2}\int_0^\infty d\lambda \frac{d\sigma_{nl}(E)}{d\Omega} . \tag{8}$$

To compare directly with the experimental results of Khayrallah *et al.* for $\sigma_*(E)$, we must calculate the cross sections for all states with $13 \leq n \leq 30$. Because of the prohibitive expense of calculating all these cross sections, we have estimated most of the cross sections contained in $\sigma_*(E)$ from the determination of some key cross sections.

From previous calculations for electron capture into low states[2] ($1 \leq n \leq 4$) we have learned that the Jackson-Schiff scaling rules[6]

$$\sigma_n(E) \equiv \sum_{l,m} \sigma_{nlm}(E) \simeq (n'/n)^3 \sigma_{n'}(E) \tag{9}$$

are accurate to within 25%. If the scaling rules hold for capture into highly excited states, we could determine $\sigma_*(E)$ from a determination of $\sigma_{13}(E)$ or even $\sigma_1(E)$. Omidvar[5] has recently questioned the validity of the Jackson-Schiff scaling rules. From an algebraic investigation of the proton-proton potential term in Eq. (1), Omidvar determined that as $n \to \infty$, the cross sections obtained by neglecting the other terms in (1) leads asymptotically to $1/n^2$ scaling for $\sigma_n(E)$. The following points remain unclear from Omidvar's presentation: (a) At about what value of n does the proton-proton term begin to dominate the asymptotic behavior of $\sigma_n(E)$? (b) Upon including the other terms in Eq. (1) does $\sigma_n(E)$ still scale as $1/n^2$? The first and second terms in the numerator cancel each other to a large extent [as can be seen from the differential cross sections $d\sigma_{nlm}(E)/d\Omega$]. It is this strong cancellation, which occurs even for large values of n, that makes a determination of $\sigma_n(E)$ from only one of the terms in Eq. (1) of questionable validity. By calculating the differential cross sections for increasing n and noting at what point this cancellation no longer appears as we go up in n values, we can determine where, if at all, approximating the cross section by considering only one term in the transition matrix element may be valid. From the present differential-cross-section calculations we must conclude that if the $1/n^2$ behavior is to be reached at all, it must be for n much larger than 15.

Omidvar used a parabolic coordinate system in his investigation so that bound Coulomb states are determined by the quantum numbers n, n_1, and $|m|$.[10] It appears that Omidvar's presentation does not hold rigorously for states with $|m|$ unequal to zero. This is due to his choice of quantization axis as being along $\hat{q}$, where $\vec{q} = \vec{C} - \vec{k}$ in Eq. (2). Since integration over the variable $\vec{k}$ in Eq. (2) is performed, the choice of the quantization axis along $\hat{q}$ does not correspond to a well-defined $|m|$ state, and therefore Omidvar's equations (7) and (8) should be modified for states with $|m|$ different from zero by including a rotational transformation $\langle nn_1 m|R|nn_1' m'\rangle$, where R is the rotation which takes the vector $\hat{q}$ into a vector along the scattering axis.

Any of the n-m wave functions in spherical polar coordinates for a fixed value of n and m (which are the n-m wave functions with different l values) can be written as a linear combination of the n-m wave functions in parabolic coordinates with fixed n and m (for fixed sign in $e^{\pm im\phi}$). Thus

$$\varphi_{nlm}(\vec{P}) = \sum_{n_1=0}^{n-m-1} a_{n_1}^{n,l,m} \varphi_{nn_1m}(\vec{P}) . \tag{10}$$

Therefore that part of the transition matrix element which comes from the proton-proton interaction for charge transfer into an nlm state (from the ground state) may be written as a linear combination of the transition matrix elements for charge transfer into nn_1m states as follows:

$$T_{nlm} = \sum_{n_1=0}^{n-m-1} (a_{n_1}^{nlm})^* T_{nn_1m} . \tag{11}$$

Omidvar obtains a selection rule for T_{nn_1m} which specifies that $T_{nn_1m} = 0$ if $m \neq 0$. This would indicate that $T_{nlm} = 0$ if $m \neq 0$, which is not the case.[11] As already mentioned, the origin of the difficulty is the quantization about the rotating $\hat{q}$ axis.

We have calculated $\sigma_n(E)$ for several high values of n to test these scaling rules by comparing them with actual calculations made feasible by the numerical method developed in II.

From the previous calculations[2] we have learned that the cross sections for higher l states are not negligible as had been assumed in the early litera-

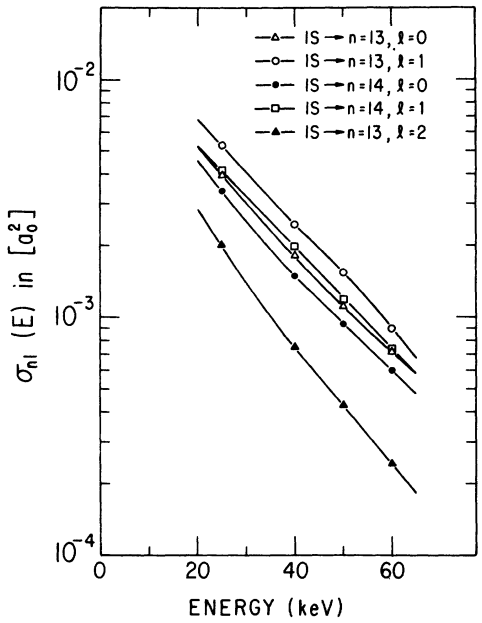

FIG. 1. Capture cross sections $\sigma_{nl}(E)$ for capture from the ground state into states n, l in units a_0^2.

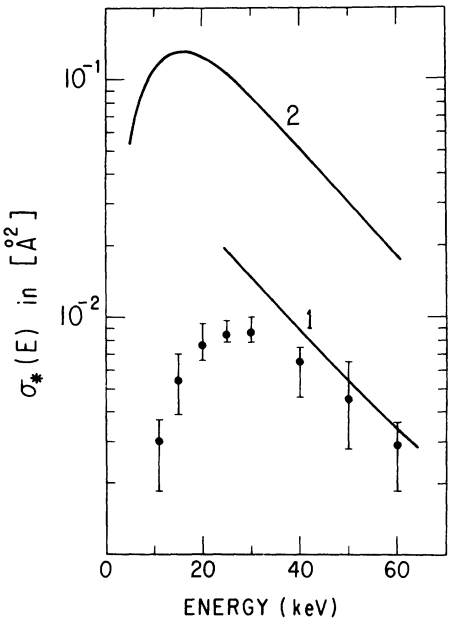

FIG. 2. Cross section for capture into all states $13 \leq n \leq 30$ in units of Å^2. (1) Present calculation; (2) results of Bulter-May-Johnston model; (Φ) data of Khayrallah, Karn, Koch, and Bayfield.

ture.[6,12] Indeed the $l=1$ cross sections can be larger than the $l=0$ cross sections for the same n, particularly for large n. However, values of $\sigma_{nl}(E)$ for $l>2$ are small compared with $l=0$, 1, and drop off rapidly with increasing l. We have also tested this trend for the highly excited n states.

We computed the cross sections $\sigma_{nl}(E)$ for the states $n=13, l=0, 1, 2$, and $n=14, l=0, 1$, at the four energies 25, 40, 50, and 60 keV, and higher l states for $n=13$ at 50 keV. The calculated differential cross sections show the same general features as obtained for the lower-n-state calculations. They fall off rapidly with increasing scattering angle, there are small oscillations at larger values of the scattering angle due to the proton-proton term in the transition matrix element, for $l=0$ cross sections there is a "dark angle" at certain values of the scattering angle where the first and second terms in the numerator of Eq. (1) cancel, the width of the dark angle region decreases with increasing energy, for $l\neq 0$ there is no dark angle because of the contribution produced by the $m\neq 0$ states to $\sigma_{nl}(E)$ at what would otherwise be the dark angle, etc.[13] The integrated cross sections are shown in Fig. 1 with the curves drawn through the calculated points. The cross sections with and without the nonorthogonality corrections are tabulated in Table I. Note that throughout the energy region calculated the $l=1$ cross sections remain larger than for $l=0$. From the calculations of $\sigma_{13,l}$ (50) keV) for $l=3, 4$ we estimate that the contribution from all states with $l\geq 2$ will amount to $\frac{1}{4}$ of $[\sigma_{n0}(E)+\sigma_{n1}(E)]$. This value will not vary much over the energy range or with changing n, for $n\geq 13$. In order to test the scaling rules we computed the ratios $\sigma_{13}(E)/\sigma_2(E)$, $\sigma_{14}(E)/\sigma_2(E)$, and $\sigma_{14}(E)/\sigma_{13}(E)$ and compared with both postulated

TABLE I. Capture cross sections in a_0^2.

	Energy (keV)							
	With orthogonality corrections				Without orthogonality corrections			
	25	40	50	60	25	40	40	60
$n=13, l=0$	3.94×10^{-3}	1.82×10^{-3}	1.10×10^{-3}	7.16×10^{-4}	3.27×10^{-3}	1.52×10^{-3}	9.26×10^{-4}	6.08×10^{-4}
$l=1$	5.28×10^{-3}	2.43×10^{-3}	1.54×10^{-3}	8.91×10^{-4}	4.73×10^{-3}	2.10×10^{-3}	1.34×10^{-3}	7.77×10^{-4}
$l=2$	2.05×10^{-3}	7.48×10^{-4}	4.27×10^{-4}	2.40×10^{-4}	2.01×10^{-3}	6.86×10^{-4}	3.84×10^{-4}	2.16×10^{-4}
$n=14, l=0$	3.33×10^{-3}	1.49×10^{-3}	9.30×10^{-4}	5.97×10^{-4}	2.78×10^{-3}	1.24×10^{-3}	7.84×10^{-4}	5.09×10^{-4}
$l=1$	4.13×10^{-3}	1.97×10^{-3}	1.19×10^{-4}	7.32×10^{-4}	3.71×10^{-3}	1.71×10^{-3}	1.03×10^{-3}	6.38×10^{-4}

rules. We found that the cross sections predicted by the Jackson-Schiff rules were 20% smaller for $\sigma_{13}(E)/\sigma_2(E)$ and $\sigma_{14}(E)/\sigma_2(E)$ and 1.5% smaller for $\sigma_{14}(E)/\sigma_{13}(E)$ than the calculated values, whereas the $1/n^2$ scaling gave results 500% larger and 7% larger for the ratios, respectively. Thus, the Jackson-Schiff scaling rules give results closer to the actual computations. It is clear that neither rule is entirely accurate. Using the Jackson-Schiff scaling we obtain the expression

$$\sigma_*(E) = \left[\sigma_{13}(E) + \sigma_{14}(E) \sum_{n=14}^{30} \left(\frac{14}{n}\right)^3\right]. \tag{12}$$

An estimate of the percentage error due to using the scaling rules and approximating the l dependence for high l is 30%. The experimental uncertainties of Khayrallah *et al.* are about 20%. Figure 2 compares the present calculated values of $\sigma_*(E)$ with the data of Khayrallah and the predictions of the Butler-May-Johnston model. The discrepancy between the present result and the experiment at 25 keV is probably due to the breakdown of the validity of our method at low energies. For the other energies, our results are within the combined experimental and theoretical error bars.

Butler, May, and Johnston use an impact-parameter method with a straight line trajectory and neglect the proton-proton interaction term. In order to sum over the bound states they use the closure property of the wave functions and subtract the continuum contributions. Their result for the total charge-exchange cross section into bound states with principle quantum numbers greater or equal to n, is

$$\sigma(\geq n) = \frac{32\pi}{5n^2} \frac{1}{E[1+(1/E)(E-\frac{1}{4})^2]^5} . \tag{13}$$

They also compute the cross sections for capture into different orbital angular-momentum components. The Butler-May-Johnston model (a) results in cross sections $\sigma(n)$, which scale as $1/n^3$ for >4, (b) decrease rapidly with $l \geq 2$, and (c) give $\sigma_{n1}(E) > \sigma_{n0}(E)$. Their model gives values of $\sigma_*(E)$ an order of magnitude larger than experiment. The model does show correctly the peaking of the cross sections at the energy where the proton velocity equals the Bohr velocity. The present method, quantitatively accurate at higher energies, breaks down completely at velocities lower than the Bohr velocity.

A more direct comparison with the present calculation can be obtained by measuring the charge transfer into only the states $13 \leq n \leq 14$. This can be accomplished[14] by isolating the peaks of $dI/d|\vec{E}|$ due to charge exchange into only the n equal 13 and 14 states, where I is the yield of excited atoms which undergo Lorentz ionization and $\vec{E}$ is the Lorentz electric field. Also, an extension of the energy range to higher energies is important to determine the asymptotic high-energy behavior of excited states since models have been proposed which lead to drastically different values for the excited state cross sections at asymptotic energies.[2]

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