Cross Sections for the Formation of Excited States in a Helium Target by the Impact of 0.15- to 1.0-MeV Protons and Deuterons. II. Comparison with Theory*

E. W. THOMAS
Georgia Institute of Technology, Atlanta, Georgia

(Received 8 June 1967)

Experimental data on the excitation of helium by the impact of protons in the energy range 0.15–1.0 MeV, published in the preceding paper, are compared with available theoretical predictions. The general predictions of the Bethe-Born approximation are examined, and empirical values are determined for the constants in the equations which will allow the prediction of the behavior to be expected at higher energies and for different projectiles. In the case of quadrupole transitions, the accuracy of available Born-approximation predictions is poor, but reasonable agreement is found with the experimental data. Accurate calculations in the distortion approximation are available for the excitation of dipole transitions, and these are found to agree very well with experiment down to 75-keV impact energy. The data for the simultaneous removal of one electron and the excitation of the remaining electron are examined qualitatively in the light of theoretical predictions and compared with other two-electron excitation processes.

I. INTRODUCTION

IN the paper immediately preceding,[1] we have reported cross sections for the formation of excited helium atoms and ions by a passage of 0.15- to 1.0-MeV protons through a neutral helium target. Cross sections were presented for the excitation of various n^1S, n^1D, and n^1P levels of neutral helium as well as an emission function for the $n=4$ to 3 transition in the helium ion. The experimental technique was to measure the apparent cross section for the emission of a spectral line by an optical method, correct for cascade and branching, using known transition probabilities, and express the data as a level excitation function. Suitable account was taken of second-order effects such as collisional transfer and absorption of resonance radiation. In the case of the helium-ion measurement, it was not practicable to make the required cascade and branching corrections due to the impossibility of separating the contributions from the states of different angular-momentum quantum number l. These data were displayed only as an emission function.

Most of the work was concerned with the direct excitation of a helium target;

$$H^+ + He \rightarrow H^+ + He(nl). \quad (1)$$

This is a particularly well-defined process from an experimental point of view since there is no ambiguity as to the post or prior states of excitation or charge of either particle. In contrast, one may consider the collisional formation of an excited helium ion which has been studied more briefly. Processes of ionization

$$H^+ + He \rightarrow H^+ + He^+(nl) + e \quad (2)$$

or charge transfer

$$H^+ + He \rightarrow H + He^+(nl) \quad (3)$$

may be responsible. A measurement of the rate of formation of the excited helium ion would, of necessity, be a sum of processes (2) and (3), with an additional ambiguity in the latter case due to possible excited states of the post-collision fast hydrogen atom.

A brief review of portions of the theory of excitation, including the general predictions of the Born and Bethe approximations, is presented in Sec. II. In Secs. III–V, the data of the preceding paper[1] on optically allowed and forbidden transitions are examined and compared with available theoretical predictions. The simultaneous removal of an electron and excitation of the remaining ion are considered in Sec. VI.

II. THE THEORY OF EXCITATION BY ION IMPACT

At sufficiently high impact energies, cross sections for direct excitation may be predicted by the Born approximation. In the case of helium, the calculations are sensitive to the wave functions employed, and discrepancies between experiment and theory at high energies will indicate that inaccurate wave functions were used. For the excitation of the ^1P levels the calculations have been further refined to include distortion, and such approximations should be expected to be accurate to somewhat lower energies than the first Born approximation. The available Born- and distortion-approximation calculations will be compared with the experimental data of the preceding paper.[1]

Of particular value in the present work is the further simplification by Bethe of the Born approximation,[2] which allows the formulation of simple general expressions predicting the asymptotic high-energy behavior of the cross section. The cross section σ_{nl} for the excitation of a single electron in the target from the ground state to some excited state nl may be written in terms of the projectile impact energy E (eV), mass M, and

* This work was partially supported by the Controlled Thermonuclear Research Program of the U. S. Atomic Energy Commission.

[1] E. W. Thomas and G. D. Bent, preceding paper, Phys. Rev. **164**, 143 (1967).

[2] N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Oxford University Press, London, 1965), pp. 497, 613.

charge Z. For the excitation of an optically allowed dipole transition,

$$\sigma_{nl}=(A_{nl}Z^2M/E)\ln(B_{nl}E/M); \quad (4)$$

and for a transition associated with a quadrupole moment but no dipole moment,

$$\sigma_{nl}=C_{nl}Z^2M/E. \quad (5)$$

The constants A_{nl}, B_{nl}, and C_{nl} are related to the dipole and quadrupole oscillator strengths, respectively. For incident protons, mass M and charge Z are both taken as unity.

The present paper examines the measured cross sections and establishes the energy region above which the energy dependence predicted by the Bethe-Born theory is exhibited. Clearly, this will also define the region in which the more accurate first Born approximation will be valid. Empirical values of A_{nl}, B_{nl}, and C_{nl} are determined from the experimental data. Since these constants are characteristic only of the target system it should be possible to use such data for extrapolation to higher energies and situations involving projectiles of different charge Z and mass M.

The Bethe-Born formulas predict that for projectiles of equal charge Z but different mass M, the cross sections should be the same at equal impact velocities, a feature which is also displayed by the results of the full Born treatment. This justifies our observation that protons and deuterons at equal velocities exhibit the same cross sections.[1] Deuterons were used to extend the lower velocity limit of some of the experimental data considered here.

III. EXCITATION OF QUADRUPOLE TRANSITIONS $1^1S\rightarrow n^1S$

The excitation of the higher n^1S states from the ground state of helium is a transition which is optically forbidden, and is expected to have the characteristics of a quadrupole transition. No calculations are available for proton-impact excitation although electron impact has been considered in the Born approximation.[3] At high velocities the Born approximation for electron and proton impact will give the same results, but at lower velocities there will be some differences due to the different ranges of applicability of certain approximations.[4] The scaling relationship suggested by Bates and Griffing[4] allows electron-impact calculations to be related to proton impact over a wide range of velocities. The results of this procedure obtained by Gaillard[5] are shown in Fig. 1, compared with the experimental measurements. Above 400-keV impact energy, the theoretical and experimental results agree within the limits of error of the experiment.

TABLE I. Experimental values of C_{nl}.

State	4^1S	5^1S	6^1S	7^1S	4^1D
$C_{nl}\times10^{-14}$	2.25	1.22	0.71	0.43	0.70

The Bethe-Born formulation of Eq. (5) is only applicable where the product of cross section and impact energy is constant. Figure 2 shows this product as a function of energy for two representative ^1S levels and indicates that the general predictions of the Bethe-Born theory are valid above 450 keV. Values of the constant C_{nl} derived from the experimental data at energies above 450 keV are listed in Table I. We observe that, within experimental error, C_{nl} varies inversely as the cube of the principal quantum number n. All the experimental data at impact energies above 450 keV where the Bethe-Born energy dependence has been demonstrated, may be summarized in a single equation:

$$\sigma=\frac{1.47\times10^{-12}}{n^3E}\ \text{cm}^2. \quad (6)$$

This gives the cross section for the excitation of the n^1S level of helium by impact of protons of energy E (eV) with an uncertainty not exceeding $\pm25\%$.

IV. EXCITATION OF THE QUADRUPOLE TRANSITION $1^1S\rightarrow 4^1D$

The calculations of the cross sections for the excitation of this quadrupole transition by McDowell and Pluta,[6] and Gaillard[5] are compared with experimental

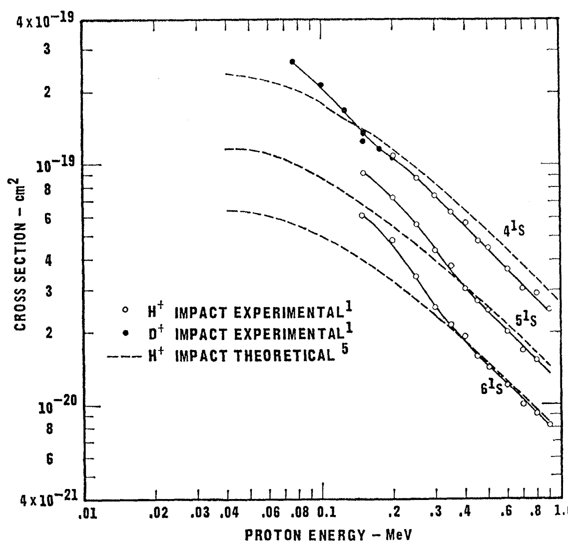

FIG. 1. Comparison of the experimental cross sections (Ref. 1) for the excitation of n^1S levels with the theoretical predictions of Gaillard (Ref. 5).

[3] M. A. Fox, Proc. Phys. Soc. (London) **86**, 789, (1965).
[4] D. R. Bates and G. Griffing, Proc. Phys. Soc. (London) **A66**, 961 (1953).
[5] M. Gaillard, Compt. Rend. **263**, 549 (1966).
[6] M. R. C. McDowell and K. M. Pluta, Proc. Phys. Soc. (London) **89**, 793 (1966).

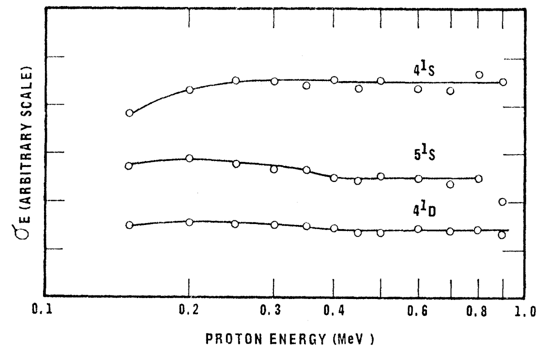

FIG. 2. The product of cross section and energy for selected levels, expressed on an arbitrary scale, as a function of energy of impact. Constancy of the product indicates validity of the Bethe-Born approximation for quadrupole transitions.

measurement in Fig. 3. The discrepancy between the theoretical calculations is due to the different choices of wave functions. McDowell and Pluta[6] quote a possible uncertainty of a factor of 2, while Gaillard[5] does not indicate the limits of accuracy of his calculations. McDowell and Stauffer[7] have previously examined various initial-state wave functions in the calculation of quadrupole strength for this transition. The most suitable wave function from the group examined gave a quadrupole strength which was considerably less than that given by the wave functions which were later employed by McDowell and Pluta[6] in the calculation of the excitation cross sections. There is no simple relationship between the quadrupole strength and the cross section for a quadrupole excitation by collision. However, a cross section calculated using the wave functions found most suitable for oscillator-strength calculations would undoubtedly give a lower cross section than that found by McDowell and Pluta, and therefore in better agreement with the experimental measurements.

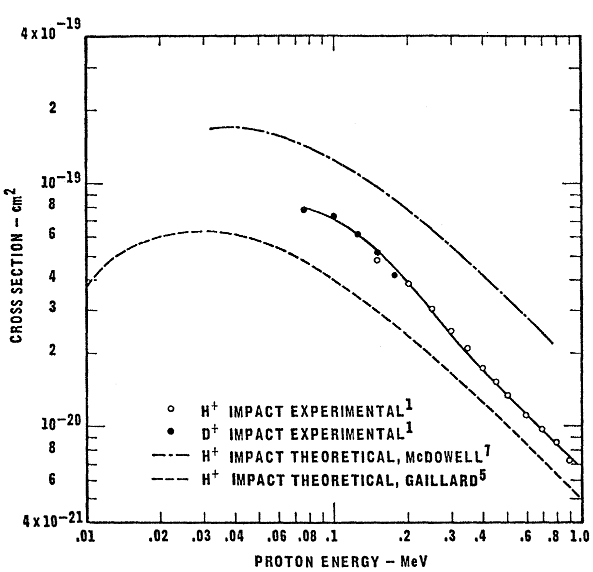

FIG. 3. Comparison of the experimental cross section (Ref. 1) for the excitation of the 4^1D level with the theoretical predictions of Gaillard (Ref. 5) and McDowell and Pluta (Ref. 6).

[7] M. R. C. McDowell and A. D. Stauffer, Phys. Letters 12, 207 (1966).

Again, in this case, it is of interest to compare the experimental data with the general predictions of the Bethe-Born approximation, as defined in Eq. (5). As shown in Fig. 2, the product of cross section and energy becomes constant above 400 keV, and the higher-energy data have been used to establish the empirical value of the constant C_{nl} which is listed in Table I.

V. EXCITATION OF THE DIPOLE TRANSITIONS $1^1S \rightarrow n^1P$

It is expected that the theoretical calculations of cross sections for the so-called "optically allowed"

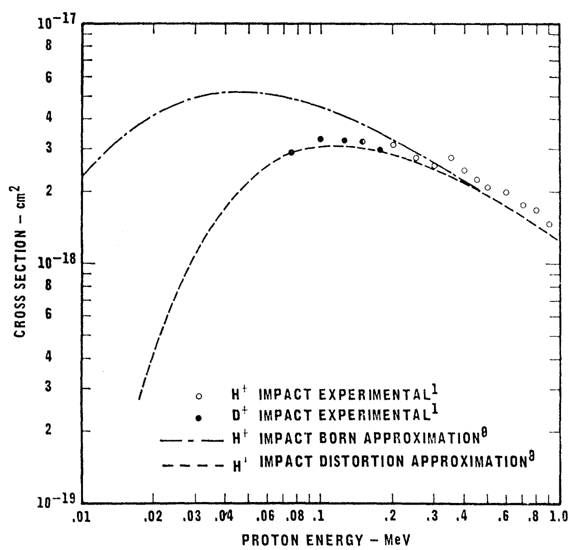

FIG. 4. Comparison of the experimental cross section for the excitation of the 3^1P level with the distortion approximation predictions by Bell (Ref. 8).

dipole transitions will be more accurate than the prediction of quadrupole transitions, since the results are less sensitive to the choice of initial-state wave functions. While quadrupole transitions have only been considered in the simple Born approximation, the calculation of dipole transitions has been refined by the inclusion of distortion. Figure 4 shows the cross section for the excitation of the 3^1P level taken from our previous paper[1] compared with the theoretical predictions of Bell,[8] both in the Born and distortion approximations. The influence of rotation coupling and back coupling on the distortion approximation for helium has not been examined in this treatment. Using the deuteron data to extend the effective velocity

[8] R. J. Bell, Proc. Phys. Soc. (London) 78, 903 (1961).

range of the experiment, good agreement between the distortion approximation and experiment is found down to 75-keV impact energy. The predictions of the Born approximation are in substantial disagreement with experiment at impact energies below 200 keV, both as regards magnitude and energy dependence. Agreement of the energy dependence of theoretical and experimental cross sections is an excellent confirmation of the predictions of the distortion approximation. The agreement of the magnitudes of the experimental and theoretical cross sections at high energies, where the simple Born approximation is applicable, suggests that the experiment is more accurate than the $\pm 35\%$ that we have estimated.[1]

The data for the excitation of the dipole transition in Fig. 4, and those for the quadrupole transitions in Figs. 1 and 3, illustrate experimentally the very considerable difference between the energy dependence of the excitation functions for the optically allowed and forbidden transitions which is predicted by the Bethe-Born approximation. The quadrupole transitions fall off very much more rapidly with increasing energy than do the dipole excitation cross sections.

An attempt was made to fit the experimental data for the excitation of the various n^1P levels to the predictions of the Bethe-Born approximation given in Eqs. (4) and (5). Surprisingly, an equation of the form $\sigma = A_{nl}E^{-1}\ln B_{nl}E$ would seem to fit all the available data through the whole energy range. However, the Bethe-Born approximation can be no more accurate than the first Born approximation. Since the present results show that the first Born approximation is not sufficiently accurate to represent all of the experimental data it would be misleading to derive empirical values of A_{nl} and B_{nl}, since they would undoubtedly have limited accuracy outside the energy range of the present work. It is clear that the energy dependence of the cross sections for the excitation of the different n^1P levels are the same within experimental error, and we identify quite accurately a dependence of the cross sections for the various levels at any one impact energy on the inverse third power of the principal quantum number.

VI. FORMATION OF THE EXCITED STATE OF THE HELIUM ION

In the preceding paper[1] we have reported the cross section for the emission of the He II $n=4 \rightarrow n=3$ (4686 Å) transition. This spectral line consists of four separate transitions between the four degenerate upper levels ($4s$, $4p$, $4d$, and $4f$) and three degenerate lower levels ($3s$, $3p$, and $3d$) which cannot be separated spectroscopically. Although it is not possible to calculate accurately the cross section for the formation of the $n=4$ level from this data, we consider it sufficiently important to make a rough estimate in a similar manner

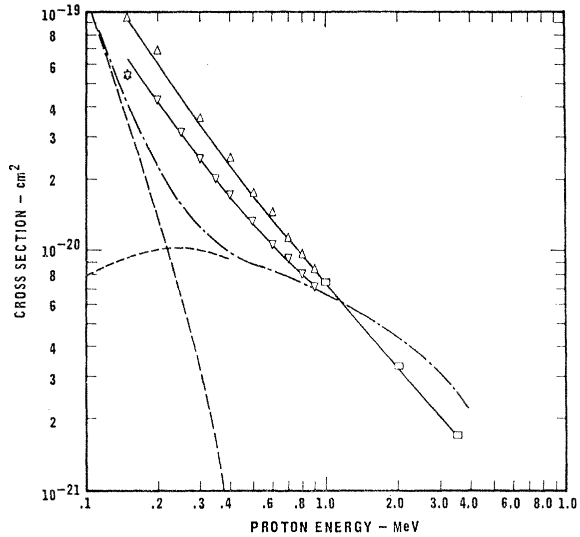

FIG. 5. Comparison of theoretical and experimental estimates of the cross section for the formation of the He^+ ($n=4$) levels by the impact of protons on a neutral helium target: — —, charge transfer process, [Eq. (3)] theoretical estimate; - - - -, ionization process [Eq. (2)] theoretical estimate; — - —, sum of theoretical estimates giving the total cross section for the formation of the He^+ ($n=4$) state by both processes; —∇—, estimate of the cross section for the formation of the He^+ ($n=4$) level from experimental data. For comparison the cross sections for the formation of the He^{2+} ion are also shown; —Δ— (Ref. 12) —□— (Ref. 13).

to Dodd and Hughes[9] and compare with the limited theoretical predictions available.

Mapelton has calculated the cross sections for the simultaneous ionization and excitation of helium[10] [Eq. (2)], and also for the simultaneous transfer of one electron to the fast incident proton and excitation of the remaining electron[11] (Eq. 3), both by the Born approximation. The charge transfer process has been calculated only for the $2s$ and $2p$ excited states, and we estimate the population of the $n=4$ states by adding these two cross sections to give a total population cross section for the $n=2$ level and scaling to the $n=4$ level, assuming that cross section is proportional to n^{-3}. Similarly, the theoretical calculation for ionization is carried out for the $n=3$ levels and we take the sum of the $3s$, $3p$, and $3d$ cross sections and again scale to give a total population of the $n=4$ state by assuming that the cross section decreases as n^{-3}. It is expected that these extrapolations should be qualitatively correct and that the absolute magnitudes should not be in error by more than a factor of 2. Figure 5 shows the cross sections for these two processes plotted separately and added together to give a total cross section for the population of the $n=4$ level. We note that the charge-transfer process predominates at low energies and reaches negligible proportions only around 300 to 400 keV.

[9] J. G. Dodd and R. H. Hughes, Phys. Rev. **135**, A618 (1964).
[10] R. A. Mapleton, Phys. Rev. **109**, 1166 (1958).
[11] R. A. Mapleton, Phys. Rev. **122**, 528 (1961).

Reference to Eq. (6) of our previous paper[1] will show that when cascade can be neglected, the cross section for the emission of the $n=4$ to $n=3$ spectral line should be proportional to the cross section for the excitation of the $n=4$ level. Mapleton's calculation of simultaneous ionization and excitation at 711-keV impact energy shows that the $3s$- and $3d$-state excitation cross sections are approximately equal and one-seventh of the $3p$ cross section. We make the assumption that the $4s$, $4p$, and $4d$ excitation cross sections are in the same ratio, and also that the $4f$ cross section is the same as the $4s$ and $4d$ levels. Using known transition probabilities and applying Eq. (6) of our previous paper,[1] we estimate that the $n=4$ level excitation cross section should be approximately a factor of 5 larger than the $n=4$ to $n=3$ emission function. Dodd and Hughes[9] also come to this same conclusion. We show our experimental estimate of the $n=4$ level population cross section derived on this basis on Fig. 5.

Although the methods by which we have arrived at our experimental and theoretical cross sections are approximate, a surprisingly good agreement is indicated in Fig. 5. We see that the energy dependence of our experimental data bears little resemblance to the separate processes of charge transfer or ionization but lies quite close to the sum of the two. The major discrepancy exists around the middle of our energy scale, in the region where the ionization process shows its maximum.

It might be expected that similarities would be found between the cross sections for the production of He^{2+} ions and excited He^{+} ions in the target. The mechanisms for both transitions may be considered as the removal of one electron by ionization or charge transfer, with the simultaneous excitation of the second electron of the target atom. In the case of the formation or excited He^{+} the second electron is excited to a group of discrete states, whereas in the formation of He^{2+} the cross section is summed over all possible excited states in the continuum. In a spirit of academic comparison we show in Fig. 5 the cross section for the production of He^{2+} ions in a helium target[12,13] compared with both the theoretical and experimental estimates of the He^{+} ($n=4$) production cross section. As we expect, there is considerable similarity between the energy dependence of the two sets of data.

VII. SUMMARY

We have taken the experimental data of a previous paper and tested available theoretical predictions. The general predictions of the Bethe-Born approximation seem to be applicable above 450 KeV, a feature which has previously been noted in the case of ionization.[14] Comparison of theory and experiment has allowed the empirical estimation of the unknown constants in the Bethe-Born approximation, and empirical formulas based on the available data are proposed for extrapolating to higher energies and excited states.

In making comparison between calculations for specific levels and the relevant experimental data, it would seem that the theoretical work may be divided into a group that has previously received experimental confirmation and a group which is relatively untested. The distortion approximation of Bell[8] shows surprisingly good agreement with experiments in the case of the 3^1P level down to the lowest impact energies used. First Born-approximation calculations derived by Gaillard[5] from the electron impact calculations of Fox[3] also show good agreement, but at high energies only, as one might expect. These particular formulations have previously been well confirmed by comparison to electron-impact work. The excitation of the ^1D levels has been predicted by the Born approximation[5,6] and relatively poor agreement is found, the theories disagreeing by a factor of 3. Neither of these formulations has been much tested by comparison with electron-impact work. In the case of the formation of the excited helium ion, comparison of the experimental data with theoretical predictions is rather difficult, but on a qualitative basis there seems to be reasonable agreement.

The preceding paper[1] compares the data from various experimental investigators. The work of Denis *et al.*,[15] and that of Robinson and Gilbody[16] is in substantial agreement with the present investigations and therefore in general accord with the conclusions drawn here. The work of Van den Bos *et al.*[17] was carried out at lower energies but clearly indicates considerably higher cross sections than those of the present investigations. The general agreement between the present experiments and the available theoretical predictions, within the estimated limitations of accuracy, give us confidence in the measurements of the previous paper.[1]

ACKNOWLEDGMENTS

The author wishes to thank L. J. Puckett and D. W. Martin for permission to use unpublished data on the formation of the He^{2+} ion by proton impact.

[12] L. J. Puckett and D. W. Martin (unpublished).
[13] S. Wexler, J. Chem. Phys. **41**, 1714 (1964); 44, 2221E (1966).

[14] J. W. Hooper, D. S. Harmer, D. W. Martin, and E. W. McDaniel, Phys. Rev. **125**, 2000 (1962).
[15] A. Denis, M. Dufay, and M. Gaillard, Compt. Rend. **264**, 440 (1967).
[16] M. Robinson and H. B. Gilbody (to be published).
[17] J. Van den Bos, G. Winter, and F. J. de Heer Physica (to be published).