# Single and double ionization of $H_2$ by electrons and protons

A. K. Edwards, R. M. Wood, and A. S. Beard
*Department of Physics and Astronomy, University of Georgia, Athens, Georgia 30602*

R. L. Ezell
*Department of Chemistry and Physics, Augusta College, Augusta, Georgia 30910*
(Received 23 November 1987)

The single- and double-ionization cross sections of $H_2$ produced by fast-electron bombardment have been measured. The electrons had selected energies between 750 and 3500 keV/amu. The results are compared to previously reported data from this laboratory for equivelocity protons on $H_2$. The ratio $R$ of double-to-single-ionization cross sections is about a factor of 2 greater for electrons than for protons. The difference occurs because of differences in both the single- and double-ionization cross sections. At 750 keV/amu, the single-ionization cross section produced by electrons is $0.83\pm0.04$ times that produced by protons. At the same velocity, the double-ionization cross section produced by electrons is $2.25\pm0.35$ times that produced by protons. The results from the $H_2$ target are compared to results from a helium target, and marked similarities are observed.

## I. INTRODUCTION

Previous studies of the double ionization of helium by electrons[1,2] and protons[3–5] showed that the double-ionization cross section produced by electrons was greater than that by equivelocity protons in the range of 0.5–5 MeV/amu. This difference was usually shown in a plot of the ratio $R$ of double-to-single-ionization cross sections. It was found that $R$ for electron collisions with helium is approximately a factor of 2 greater than it is for proton collisions. More recently, it was shown[6] that antiproton collisions with helium yield values of $R$ nearly the same as those with electrons.

McGuire[7] has suggested that this difference is due to the interference of scattering amplitudes for two processes. One process involves a direct double collision where the projectile interacts separately with each target electron to produce double ionization. The other process is a rearrangement or shakeoff process where the projectile interacts with a single electron to remove it, and the second electron is removed when the ion relaxes to a continuum state. According to McGuire, the latter process is described by a first Born term with a scattering amplitude proportional to $-Z$ of the projectile ($Z$ being the projectile charge), while the former process is described by a second Born term that is proportional to $Z^2$ of the projectile. The cross term of the amplitudes that occurs in the calculation of the cross section then either increases or decreases the cross section depending upon whether the projectile charge is negative or positive.

Reading and Ford[8] discuss an argument attributed to Becker that disputes the McGuire model. The argument is that (1) The direct double-collision process leads to two $p$-state electrons in the final state due to the dipole nature of the interaction; (2) the shakeoff process leads to one $p$-state electron and one $s$-state electron in the final state because of the monopole nature of the shakeoff process;[9] and (3) the two processes cannot interfere because of the different final states, and therefore, there will be no $Z^3$ term in the cross section.

In the same paper, Reading and Ford[8] report a forced-impulse calculation of the double ionization of helium by fast projectiles. They express the double-ionization cross section for 2.31-MeV/amu projectiles on helium in a Born-series expansion in the charge of the projectile. The difference between $p^+$ and $p^-$ projectiles is shown to occur because of a $Z^3$ term in the series that arises from nondipole interactions between the projectile and target electrons. The calculations account reasonably well for the observed differences in double ionization produced by protons, antiprotons, and electrons.

In order to picture the full collision process, the authors propose what they call the interception model, which is based on the role of final-state correlations between the ionized electrons. In the interception model, one electron is ionized and begins to move out of the atom. Its motion is intercepted by the second electron, and the interaction causes both to leave the atom. The first and second Born amplitudes needed to yield a $Z^3$ interference term in the cross section result from either one or two interactions with the same electron. Double ionization results from electron-electron interaction in the final state. The difference between the proton and antiproton data occurs because the oppositely charged projectiles modify the target electron distribution and cause different correlations in the final state.

The interception model is supported by the classical-trajectory Monte Carlo (CTMC) calculations of Olson[10] for large impact parameters with $p^-$ projectiles and small impact parameters with $p^+$ projectiles. The first electron interacts with the projectile and follows a curved path around the nucleus and interacts with the second electron. In addition to interception, the calculations of Olson show that at small impact parameters with $p^-$ screening of the nucleus occurs that enhances double ionization.

Vegh[11] has proposed a similar polarization model. Vegh assumes that the projectile interacts once with each target electron. The separate interactions are related by the correlations between the electrons. When the projectile is a proton, one electron is pulled closer to the projectile and the other is relatively unaffected; the result is that the projectile is less likely to encounter both electrons. On the other hand, when the projectile is an antiproton, the projectile-electron repulsion drives the two electrons closer together; the result is that the projectile is more likely to encounter both electrons. Vegh reports satisfactory agreement between his independent-particle model calculations and the proton and antiproton data.

It has been suggested[12] that the difference in the double-ionization cross sections between equivelocity electrons and protons might be due to the mass differences of the two types of projectiles. However, the antiproton measurements[6] match those of electrons and indicate that it is the projectile charge and not its mass that is the important factor.

As mentioned earlier, comparisons of double-ionization cross sections caused by protons and electrons have often been presented as measurements or calculations of the ratio of double-to-single ionization. In those comparisons it is usually assumed that the single-ionization cross sections produced by equivelocity electrons and protons are equal, and that any differences in the ratio $R$ arise because of differences in the double-ionization cross sections. Indeed, if the various measurements of the single ionization of helium made at different laboratories are compared, a wide range of values for $\sigma^{+}(e^{-})/\sigma^{+}(H^{+})$ is found. The value is close to one, but the uncertainty is large. To remove uncertainties, we have made measurements of both cross sections with the same apparatus and the same experimental conditions. We find that the ratio is not unity at all energies, and show the influence of this observation on calculations of the ratio $R$ and double-ionization cross sections. The work of Olson[10] also shows that single ionization does not follow a simple $Z^2$ scaling and single ionization of helium by protons below 1 MeV is greater than that by antiprotons. The enhancement is due to saddle-point electrons that can occur for the positive projectile.

The present work concentrates on the hydrogen molecule rather than the helium atom as a target. The hydrogen molecule and helium are both two-electron systems, although the electron distributions are decidedly different. In each case, however, double ionization must involve both electrons. In our report, we compare the behavior of the two targets when bombarded by electrons and protons.

The helium atom has spherical symmetry in the ground state whereas $H^2$ has $D_{\infty h}$ symmetry. Because of the symmetry of the molecule, we are able to measure the double-ionization cross section of $H_2$ as a function of the orientation of the internuclear axis of the molecule. This is true because the molecular rotation times are long compared to the dissociation times. This point has been discussed in earlier work.[13]

## II. EXPERIMENTAL PROCEDURE

The experimental procedure used in the double-ionization measurements is illustrated in Fig. 1, and is described in more detail in earlier reports.[14] A pulsed beam of electrons or protons passes through a differentially pumped collision region containing the $H_2$ target gas and is then collected in a Faraday cup. The beam pulses are approximately 200 ns in duration, and are separated by intervals of tens of microseconds. The target-gas pres-

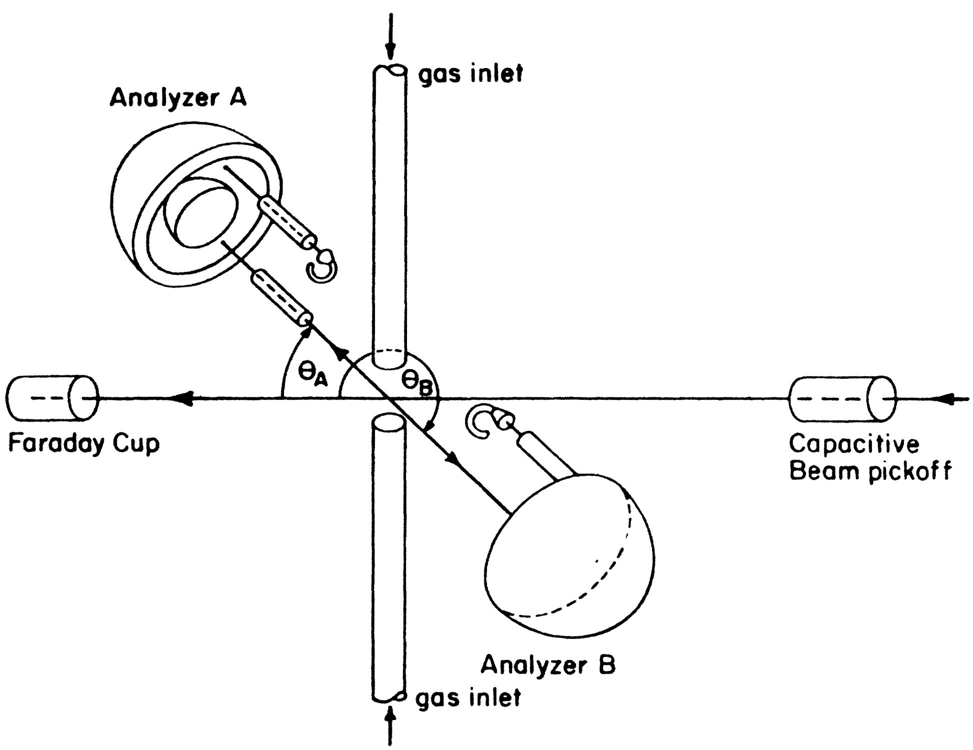


FIG. 1. Schematic of the apparatus used to measure the double-ionization cross section of $H_2$ by fast projectiles.

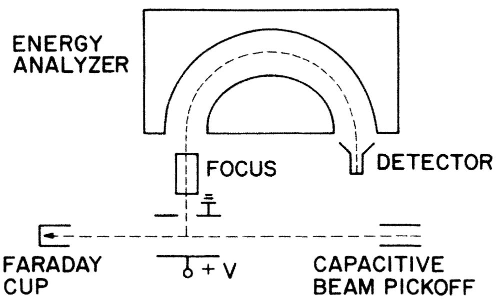


FIG. 2. Schematic of the apparatus used to measure the single-ionization cross section of $H_2$ by fast projectiles.

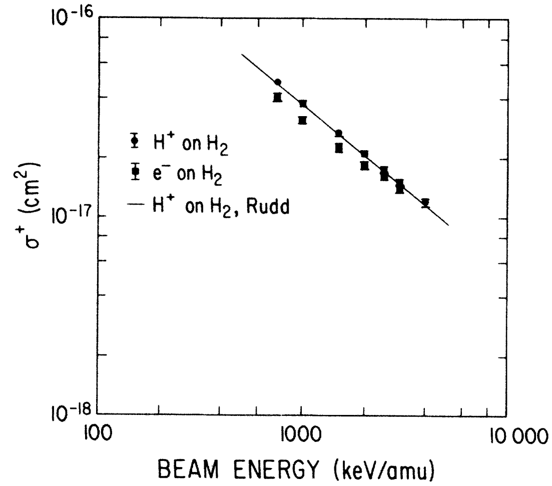


FIG. 4. Single-ionization cross sections of $H_2$ by electron and proton bombardment. The solid line shows the recommended values of Rudd *et al.* (Ref. 15) for the cross sections for electron production by protons on $H_2$.

sure is approximately 1 mTorr. Two hemispherical analyzers are positioned on opposite sides of the target in the horizontal plane. They can be independently rotated about a vertical axis through the target. The analyzers and associated instruments are arranged to measure both the kinetic energy and the time of flight of the detected ions. The dual measurements permit isolation of the $H^+$ ions of interest in this work from background ions of differing mass to charge ratio.

When both electrons are removed from the $H_2$ target, the remaining $H^+$ fragments separate from each other along a straight line with about 10 eV of energy each. These two fragments are recorded in coincidence to determine that double ionization has occurred. The analyzer voltages are adjusted to maximize the coincidence counting rate. This procedure insured that the apparatus is tuned to examine the peak of the double-ionization dissociation channel. The accidental-coincidence counting rate is negligible, amounting to much less than one percent of the full coincidence rate.

The experimental procedure used to measure the nondissociative single-ionization cross sections is illustrated in Fig. 2. In this procedure, one of the analyzers is removed, and a pair of parallel plates is placed in the collision region. The plates are biased with a potential difference of 10 V to accelerate the $H_2{}^+$ ions toward the remaining analyzer. The low accelerating voltage is used in order to minimize the deflection of the primary electron beam. The projectile beam is approximately 1 mm in diameter in the collision region. As a consequence, $H_2{}^+$ ions are formed at different points in the accelerating field (about 0.5 V/mm), and enter the analyzer with different energies. A small sweep voltage is placed on the electrostatic analyzer in order to detect $H_2{}^+$ ions over the small distribution of energies involved.

Because the energy of an $H_2{}^+$ ion is determined by its point of origin in the applied electric field, the $H_2{}^+$ energy spectrum measured by the electrostatic analyzer is correlated with the geometrical profile of the primary projectile beam. The position and width of the electron and proton beams in the collision region can be deduced directly from the observed energy spectrum. One of the objectives of this work is to make an accurate comparison of the electron- and proton-induced single-ionization cross sections. To accomplish this aim it is necessary that the projectile beam position and profile be the same

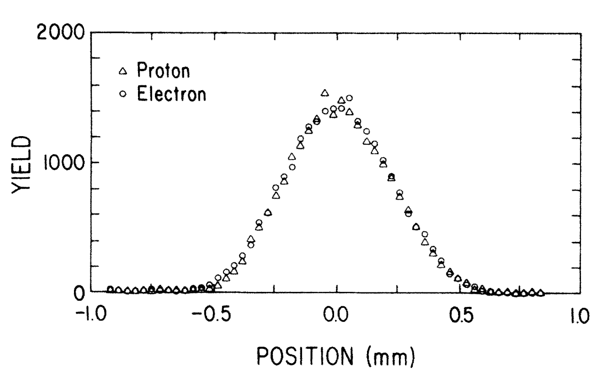


FIG. 3. Profiles of the proton and electron beams used in the ionization cross section measurements.

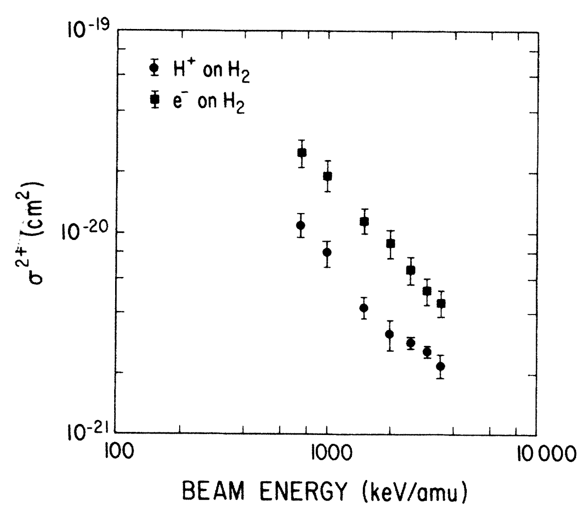


FIG. 5. Double-ionization cross sections of $H_2$ by electron and proton bombardment.

TABLE I. Single- and double-ionization cross sections for $e^-$ on $H_2$ and the ratio of double-to-single-ionization cross sections.

| Energy (eV) | Energy (keV/amu) | $\sigma^+$ ($10^{-17}$ cm$^2$) | $\sigma^{2+}$ ($10^{-20}$ cm$^2$) | $R$ ($10^{-4}$) |
|---|---|---|---|---|
| 408 | 750 | 4.05±0.21 | 2.48±0.39 | 6.12±0.43 |
| 545 | 1000 | 3.07±0.11 | 1.89±0.30 | 6.16±0.43 |
| 817 | 1500 | 2.26±0.10 | 1.15±0.18 | 5.09±0.36 |
| 1089 | 2000 | 1.84±0.07 | 0.891±0.139 | 4.84±0.34 |
| 1362 | 2500 | 1.63±0.06 | 0.660±0.103 | 4.05±0.28 |
| 1638 | 3000 | 1.40±0.07 | 0.520±0.081 | 3.71±0.26 |
| 1906 | 3500 | 1.19±0.06 | 0.450±0.070 | 3.78±0.26 |

for both kinds of projectiles. A small aperture serving as an exit window to define the beam profile in the collision region is placed downstream from the last focusing element of the electron gun. When changing from electron to proton bombardment, the cathode of the gun is removed, but all other apertures are left in place. The proton beam passes along exactly the same path as the electron beam. Figure 3 shows the measured profiles of the projectile beams used in the ionization measurements. As can be seen, the width of the beams are about 1 mm at their bases, and the overlap is excellent.

The apparatus used in the single-ionization measurements was calibrated by using helium as a target gas and measuring the single ionization of helium by protons (0.75–3.0 MeV/amu) and normalizing the results to the recommended values of Rudd *et al.*[15] In earlier work[14] we normalized our results to the values obtained by Shah and Gilbody[16] which were slightly above those of Rudd *et al.*

The relative cross sections for the double ionization of $H_2$ by protons was made earlier in our laboratory.[14] To place the measurements on an absolute scale, estimates of gas pressure, detector efficiencies, and detection geometry were required. There were no absolute measurements with which to compare. The value of the ratio $R$ for protons or electrons is directly affected by the scaling procedure, but the comparison of the electron data to the proton data should not be influenced. The data for the double ionization of $H_2$ by electrons were normalized to the proton results by comparing the respective number of coincident events per integrated beam current at 750 keV/amu. At this energy, charge capture by protons is negligible.

## III. RESULTS

Figures 4 and 5 show the single- and double-ionization cross sections of $H_2$ bombarded by protons and electrons. Table I lists the values of the cross sections and their ratios for electron bombardment.

The ratio of single-ionization cross sections for electron bombardment to proton bombardment is shown in Fig. 6 and the values listed in Table II. According to the first Born approximation, this ratio is expected to be one. As can be seen, it is not in this bombarding energy region. Olson[10] has done a classical trajectory Monte Carlo calculation of the single-ionization cross sections for 750 keV/amu electrons, positrons, protons, and antiprotons on helium. He found the ratio of the cross sections for electrons to protons is 0.935±0.034; the ratio for positrons to protons is 1.005±0.037; the ratio for antiprotons to protons is 0.946±0.035. We have measured the single-ionization cross sections for both protons and electrons on helium, and find the ratio to be the same within errors as those listed in Table II. The Olson results and our measurements indicate a small effect on the single-ionization cross section due to the sign of the projectile charge.

Also shown in Fig. 6 and listed in Table II are the ratios of double-ionization cross sections for electrons and protons on $H_2$. The factor of 2 that occurs for a helium target also appears for the $H_2$ target. The ratio $R$ of

TABLE II. Single-ionization cross sections for $H^+$ on $H_2$ and the ratio of single-to-single-ionization cross sections and double-to-double-ionization cross sections for equivelocity $e^-$ and $H^+$ on $H_2$.

| Energy (eV) $e^-$ | Energy (keV/amu) $H^+$ ($D^+$) | $\sigma^+(p^+)$ ($10^{-17}$ cm$^2$) | $\sigma^+(e^-)/\sigma^+(p^+)$ | $\sigma^{2+}(e^-)/\sigma^{2+}(p^+)$ |
|---|---|---|---|---|
| 408 | 750 | 4.86±0.17 | 0.83±0.04 | 2.25±0.35 |
| 545 | 1000 | 3.77±0.13 | 0.81±0.04 | 2.37±0.37 |
| 817 | 1500 | 2.71±0.09 | 0.83±0.04 | 2.69±0.42 |
| 1089 | 2000 | 2.11±0.07 | 0.87±0.04 | 2.82±0.44 |
| 1362 | 2500 | 1.75±0.06 | 0.93±0.05 | 2.32±0.36 |
| 1638 | 3000 | 1.49±0.05 | 0.94±0.05 | 2.02±0.32 |
| 1906 | 3500 | | | 2.05±0.32 |

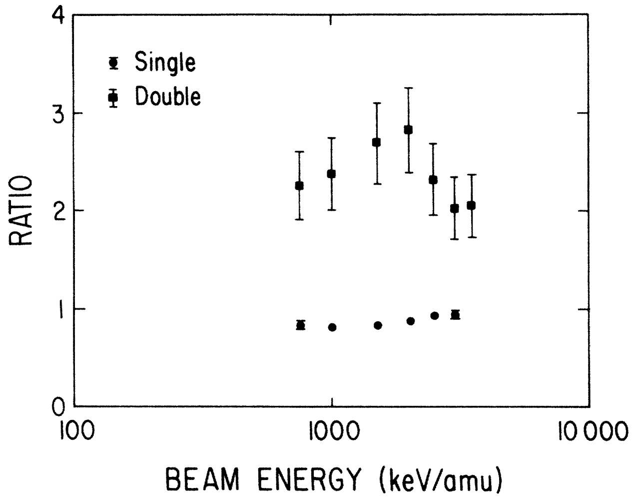


FIG. 6. The ratios $\sigma^{+}(e^{-})/\sigma^{+}(p^{+})$ and $\sigma^{2+}(e^{-})/\sigma^{2+}(p^{+})$ for electron and proton bombardment of $H_2$.

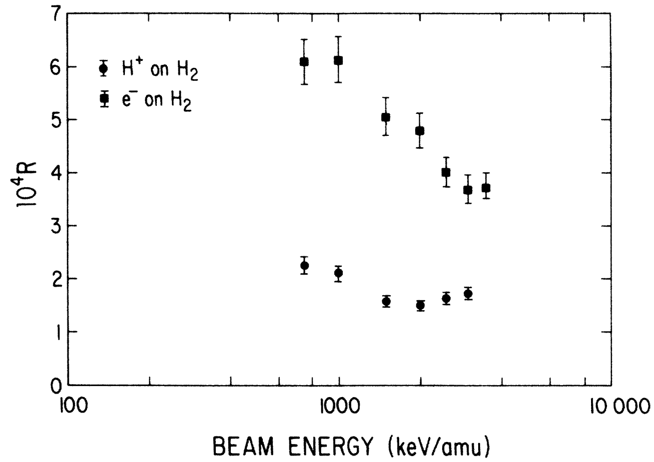


FIG. 7. The ratios $\sigma^{2+}/\sigma^{+}$ for electrons on $H_2$ and protons on $H_2$.

double-to-single-ionization cross sections for a given projectile is plotted in Fig. 7. The ratio shows the same general features as in the case of the helium target in this energy range, but $R$ is roughly a factor of 10 smaller for the $H_2$ target than it is for the helium target. A possible reason for this difference is that the electrons in the $H_2$ molecule are farther apart than they are in the helium atom ($1.7a_0$ compared to $1.0a_0$). This tends to reduce electron correlations in $H_2$ and make direct double collisions less likely.

Measurements of the effect of molecular orientation on the double-ionization cross sections showed no effect. Coincident events were recorded for the electron beam measurements with the two analyzers set at 90° relative to the beam axis and at 45° relative to the beam axis. No statistically meaningful differences were observed, and the two sets of data were averaged.

## ACKNOWLEDGMENT

This research was supported by the National Science Foundation under Grant No. PHY-8506599.

[1] B. L. Schram, A. J. H. Boerboom, and J. Kistemaker, Physica **32**, 185 (1966).
[2] P. Nagy, A. Skutlartz, and V. Schmidt, J. Phys. B **13**, 1249 (1980).
[3] S. Wexler, J. Chem. Phys. **41**, 1714 (1964); **44**, 2221 (1966).
[4] H. Knudsen, L. H. Andersen, P. Hvelplund, G. Astner, H. Cederquist, H. Danared, L. Liljeby, and K.-G. Rensfelt, J. Phys. B **17**, 3545 (1984).
[5] M. B. Shah and H. B. Gilbody, J. Phys. B **18**, 899 (1985).
[6] L. H. Andersen, P. Hvelplund, H. Knudsen, S. P. Moller, K. Elsener, K.-G. Rensfelt, and E. Uggerhoj, Phys. Rev. Lett. **57**, 2147 (1986).
[7] J. H. McGuire, Phys. Rev. Lett. **49**, 1153 (1982).
[8] J. F. Reading and A. L. Ford, J. Phys. B **20**, 3747 (1987).
[9] T. A. Carlson, Phys. Rev. **156**, 142 (1967).
[10] R. E. Olson, Phys. Rev. A **36**, 1519 (1987); (private communication).
[11] L. Vegh, *Abstracts of Contributed Papers, Proceedings of the Fifteenth International Conference on the Physics of Electronic and Atomic Collisions, Brighton, United Kingdom, 1987,* edited by J. Geddes, H. B. Gilbody, A. E. Kingston, C. J. Latimer, and H. J. R. Walters (Queen's University, Belfast, 1987), p. 451.
[12] J. F. Reading, Bull. Am. Phys. Soc. **29**, 821 (1984).
[13] R. L. Ezell, A. K. Edwards, and R. M. Wood, J. Chem. Phys. **81**, 1341 (1984).
[14] A. K. Edwards, R. M. Wood, and R. L. Ezell, Phys. Rev. A **31**, 99 (1985); **34**, 4411 (1986).
[15] M. E. Rudd, Y.-K. Kim, D. H. Madison, and J. W. Gallagher, Rev. Mod. Phys. **57**, 965 (1985).
[16] M. B. Shah and H. B. Gilbody, J. Phys. B **15**, 3441 (1982).