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Lyman-Alpha Production in H^+-$H(1s)$ Collisions*

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Lyman-alpha production in collisions between protons and hydrogen atoms has been measured in a crossed beam experiment, within the energy range 600 eV to 30 keV. The Lyman-alpha photons result both from excitation of thermal target atoms—$H^+ + H(1s) \rightarrow H^+ + H(2p)$—and from electron capture into excited states of the projectiles—$\mathbf{H^+} + H(1s) \rightarrow \mathbf{H}(2p) + H^+$. These two processes are experimentally distinguishable in terms of the Doppler shift in the capture radiation. Measurements of the angular distribution of the Lyman-alpha photons, made with an oxygen-filtered iodine-filled counter, enable the signals from the two processes to be separately determined. Relative cross sections so obtained are normalized with the aid of earlier measurements of Lyman-alpha production in e^--$H(1s)$ collisions. The results are discussed in the light of recent theoretical developments.

INTRODUCTION

DESPITE the vast amount of information accumulated over the past few years, there has been remarkably little common ground between experiment and theory concerning processes involving the transfer of an electron between a positive ion and a neutral particle. In general, the most precise theoretical treatments[1] have been confined to one-electron problems which are quite difficult to study experimentally. A small number of theoretical treatments of more complex systems have been carried out, but comparison between experiment and theory here is of limited value because of the approximate nature of the theory and the fact that the experiments rarely provide information on the states of all the interacting particles.

It has become increasingly important from a theoretical standpoint that experiments be conducted upon systems which are susceptible to the most accurate theoretical treatment. Accordingly, experimental investigation within the energy range 600 to 30 000 eV has been carried out for the processes:

$$\mathbf{H^+} + H(1s) \rightarrow \mathbf{H^+} + H(2p) \qquad (1)$$

$$\rightarrow \mathbf{H(2p)} + H^+, \qquad (2)$$

where the energetic particles are represented by boldface type.

APPARATUS AND PROCEDURE

The basic apparatus as it pertains to the production of the proton and H-atom beams is essentially unchanged from that described elsewhere.[2] The pertinent features are shown in Figs. 1 and 2.

Ground-state hydrogen atoms issuing from a tungsten furnace, heated to about 2700°K in the first of three separately pumped vacuum chambers, are collimated as they pass through the intermediate chamber, which acts as a vacuum buffer. On entering the third chamber in which the experiment is performed, the atom beam is modulated at 100 cps by a rotating toothed chopper wheel before arrival at the interaction region.

Ions extracted from a Duoplasmatron source are focused with an *einzel* lens before entering an analyzing magnetic field which selects the proton component for transmission through a series of collimating apertures before passage through the neutral beam and collection in a Faraday cup.

The region of interaction of the two beams is viewed by an ultraviolet photon counter[3] which can be rotated about the neutral beam axis in the plane containing the ion beam. The output of the counter is fed through a quenching circuit to a pulse shaper and then through a 100 cps amplifier and phase-sensitive detector.

The counter itself is sensitive to radiation within the wavelength range 1080 to 1260 Å. However, in transit to the counter, the radiation passes through a cell containing dry molecular oxygen which is opaque in this wavelength region except at seven extremely narrow transmission windows.[4] The over-all detection system is therefore responsive only to radiation at the wavelengths of these windows, one of which corresponds precisely with that of Lyman-alpha radiation (1215.7 Å). The other windows appear at wavelengths which do not correspond with photons produced in H^+-H collisions.

Lyman-alpha photons are produced in H^+-H collisions both from excitation of the thermal H atoms [reaction (1)] and from electron capture into an excited state of the hydrogen atom [reaction(2)]. The two mechanisms are distinguishable by virtue of the fact that in the vast majority of collisions, negligible momentum is exchanged between the colliding particles and the first process gives rise to thermal-energy $2p$ atoms, while the second process leads to excited atoms

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[1] D. R. Bates, in *Atomic and Molecular Processes*, edited by D. R. Bates (Academic Press Inc., New York, 1962), p. 549.

[2] W. L. Fite, A. C. H. Smith, and R. F. Stebbings, Proc. Roy. Soc. (London) **A268**, 527 (1962).

[3] R. T. Brackmann, W. L. Fite, and K. E. Hagen, Rev. Sci. Instr. **29**, 125 (1958).

[4] K. Watanabe, in *Advances in Geophysics* (Academic Press Inc., New York, 1958), Vol. V, p. 153.

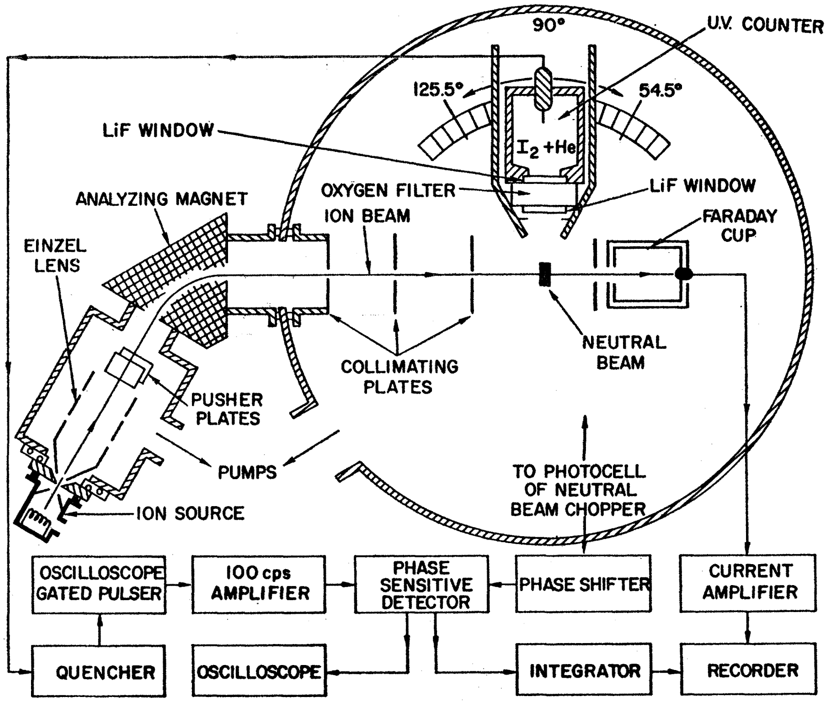

FIG. 1. Schematic diagram of the apparatus.

having the same kinetic energy as the initial protons. At each value of the primary ion energy, measurements of the Lyman-alpha intensity at two angular positions of the counter are needed in order to capitalize on this distinction and to determine the cross sections for each process. The two angles selected for these measurements are 90 and 54.5 deg with respect to the ion beam (see Fig. 1).

(a) In the 90 deg position, the counter axis is perpendicular to the plane containing the two beams. Both excitation and capture contribute to the counter signal in this position.

(b) In the 54.5 deg position, the velocity component of the projectiles along the viewing direction causes the wavelength of the Lyman-alpha radiation, resulting from capture, to be Doppler shifted by an amount which is sufficient to ensure almost total attenuation in the oxygen cell except at energies below 3 keV. In this position, the detector responds only to the unshifted Lyman-alpha radiation produced via reaction (1).

A number of corrections, which are discussed in the next section, are applied to these data in order to obtain relative cross sections for reactions (1) and (2). These cross sections are then normalized with the aid of measurements of Lyman-alpha production in electron-hydrogen atom collisions. For this purpose, the photon counter is moved to view the region of interaction of the same neutral H-atom beam with an electron beam as shown in Fig. 2. Measurements at the two counter positions (see Fig. 2), with allowance for the neutral beam divergence, enable the ratio of the cross sections for Lyman-alpha production in e^--H and H$^+$-H collisions to be determined. Knowing the absolute e^--H cross section,[5] taken to be 0.37 πa_0^2 at 300 eV, the absolute H$^+$-H cross section was determined at 5 and 10 keV. The cross sections at all other energies were then determined relative to these values.

Occurring concurrently with the processes under investigation were reactions involving excitation and capture to the 2s state of atomic hydrogen. These processes have not been investigated experimentally, although some theoretical calculations carried out by Lovell and McElroy[6] show that the capture cross section may be greater than 10^{-17} cm^2 over much of the energy range of the present experiments, while the excitation cross section is somewhat lower. The unperturbed lifetime of H(2s) atoms is greater than a few milliseconds[7] but, in the presence of an electric field, the lifetime[8] is given by

$$t=(1/2780F^2)\mathrm{sec},$$

where F is the applied electric field in V/cm. To prevent possible field quenching of 2s atoms with consequent emission of Lyman-alpha radiation, considerable care was taken to eliminate stray electric fields at the interaction region. Gas quenching[7] of the 2s atoms is negligible at the residual pressure in the experimental chamber.

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[5] W. L. Fite and R. T. Brackmann, Phys. Rev. **112**, 1151 (1958); and W. L. Fite, R. F. Stebbings, and R. T. Brackmann, *ibid.* **116**, 356 (1959).

[6] S. E. Lovell and M. B. McElroy, Kitt Peak National Observatory Report, Tucson, Arizona, 1957 (unpublished).

[7] W. L. Fite, R. T. Brackmann, D. G. Hummer, and R. F. Stebbings, Phys. Rev. **116**, 363 (1959); (E) **124**, 2051 (1961).

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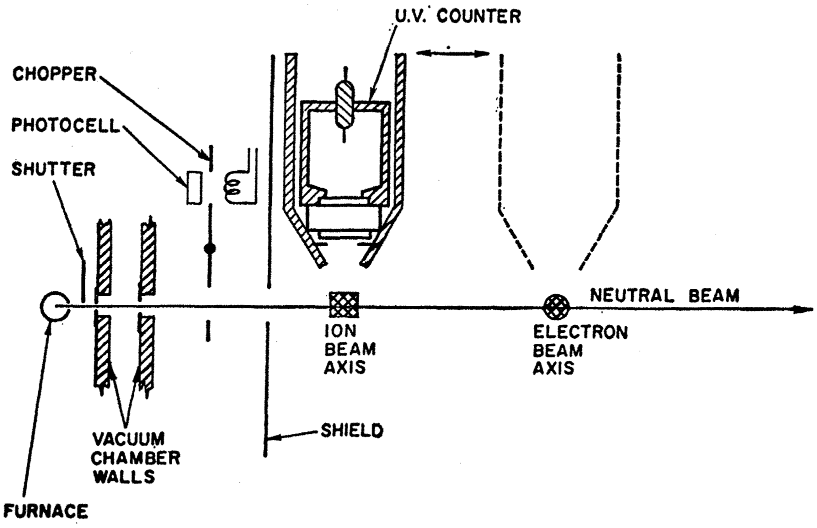

FIG. 2. Schematic diagram of the apparatus. The counter is shown in position for the H^+-H measurements. The dashed lines show position for e^--H measurements.

The production of Lyman-alpha radiation at surfaces bombarded by protons has been shown to be quite efficient[9] and care was taken to ensure that no surfaces on which primary or secondary ions would impinge were in the photon counter field of view.

Lyman-alpha production resulting from impact of the primary protons with undissociated hydrogen in the neutral beam was minimized by operation of the neutral beam source under conditions of high dissociation. Using the results of Dunn *et al.*[9] for Lyman-alpha production in H^+-H_2 collisions, it is estimated that the contribution to the observed signal introduced in this manner is less than the scatter of the experimental data.

RESULTS AND DISCUSSION

In deriving the cross sections for reactions (1) and (2) from the raw experimental data, corrections were made for:

Lost Counts. A correction must be applied for those unobserved events which occur during the dead time following a count. The corrected count rate n_T is given by

$$n_T = n_m/(1-n_m\tau)\,, \tag{3}$$

where n_m is the observed count rate and τ is the recovery time of the counter (~500 μsec). In practice, the observed count rate was invariably below 10 000 cpm and, therefore, $n_m/n_T \geq 0.9$.

Doppler Effects. The signal observed at 54.5 deg arises principally from excitation. However, at the lowest ion energies, the Doppler-shifted radiation resulting from electron capture is not totally absorbed in the oxygen filter. The wavelength shift $\Delta\lambda$ at this angular position is given by

$$\frac{\Delta\lambda}{\lambda} = v \cos 54.5°/c\,, \tag{4}$$

where v is the particle velocity, c is the velocity of light, and λ is the wavelength of Lyman-α. Thus,

$$\Delta\lambda = 1.03\sqrt{V}\,, \tag{5}$$

where $\Delta\lambda$ is in angstrom units when V is the ion energy in keV. The attenuation experienced in the filter is determined using the data of Watanabe,[4] who observed a pronounced pressure dependence of the absorption coefficient (k) of O_2 at Lyman-α due possibly to photochemical decomposition. At the O_2 cell pressure used in the present measurements (~1000 mm Hg), an extrapolated value of $k = 0.80$ cm^{-1} is obtained from their data. The absorption coefficients at adjacent wavelengths are then obtained using the profile data for the O_2 window at Lyman-α and assuming the same pressure dependence of the absorption coefficient.

It is found that at proton energies above about 2.5 keV, the Doppler shift is sufficiently large to ensure negligible contribution to the 54.5-deg signal from capture. At 600 eV, the lowest energy at which measurements were made, the capture process is estimated to contribute about 15% of the signal observed at 54.5 deg.

Polarization Effects. As a general rule, when a gas is excited by a particle beam, the emitted radiation has a nonuniform angular distribution which is related simply to the percentage polarization P of the radiation emitted perpendicular to the beam.[10] For dipole radiation, the angular distribution of the radiation intensity is given by

$$I(\theta) = (3I_0/4\pi(3-P))(1-P\cos^2\theta)\,, \tag{6}$$

where $I(\theta)$ is the intensity per unit solid angle emitted in the direction between θ and $\theta + d\theta$ with respect to the charged particle beam, and I_0 is the total intensity. For observation at 54.5 and 125.5 deg, $I(\theta)$ is independent of the polarization and is given by

$$I(\theta) = I_0/4\pi\,. \tag{7}$$

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[9] Gordon H. Dunn, Ronald Geballe, and Donavon Pretzer, Phys. Rev. **128**, 2200 (1962).

[10] L. C. Percival and M. J. Seaton, Phil. Trans. Roy. Soc. London **A251**, 113 (1958).

The choice of viewing angle in the present measurements was dictated by this consideration. Thus, regardless of the fact that the polarization may differ for proton and electron impact, the ratio of the total excitation cross sections for ion and electron impact may be obtained from measurements at 54.5 deg.

The signal observed at 90 deg includes contributions from capture and excitation. The capture cross section may be obtained from this signal simply by subtraction of the contribution due to excitation. The excitation data obtained at 54.5 deg may be used for this purpose provided account is correctly taken of the anisotropy of the excitation radiation. Since it was not possible to determine the polarization of the excitation radiation in the present measurements, recourse was taken to the work of Fennema[11] who has obtained an expression for the polarization of impact radiation in H^+-H collisions, of the form

$$P=(1-x)/(a+bx)\,, \tag{8}$$

where a and b are constants and $x=(\sigma_{\pm 1})/\sigma_0$. Here ± 1 and 0 refer to the values of the magnetic quantum number n, defined with respect to the axis of symmetry. The σ's are the corresponding cross sections for excitation to these states. Using Eq. (6) and these values for P, which are a function of the mass and energy of the colliding particle, the correction is calculated. For proton impact,[11] the polarization varies from -0.03 at 30 keV to $+0.23$ at 600 eV and the correction is therefore always less than 10%.

H(2p) Lifetime Effect. The finite lifetime[8] of the $2p$ state (1.595×10^{-9} sec), enables some of the fast excited atoms to escape from the field of view of the photon counter before decay to the ground state. It is estimated that in the worst case, i.e., at 30 keV where the atomic velocity is $\sim 2.4\times 10^8$ cm sec^{-1}, about 26% of the fast atoms decay outside the counter field of view.

To allow for these foregoing effects, the cross sections for excitation (σ_E) and capture (σ_C) are obtained from

$$\sigma_E=Q_{54.5}-\sigma_C a(1-b) \tag{9}$$

and

$$\sigma_C=\frac{Q_{90}-RQ_{54.5}}{(1-aR)(1-b)}\,, \tag{10}$$

where $Q_{54.5}$ and Q_{90} are the phenomenological cross sections determined directly from the signals observed at 54.5 and 90 deg., a is the ratio of the detection efficiency for capture and excitation radiation emitted at 54.5 deg., b is the fraction of the fast $2p$ atoms which decay outside the field of view of the counter, and R is the ratio of the intensities of the excitation radiation emitted at 90 and 54.5 deg. Both cross sections may include some contribution from cascade, and σ_C is evaluated on the assumption that the capture radiation is unpolarized.

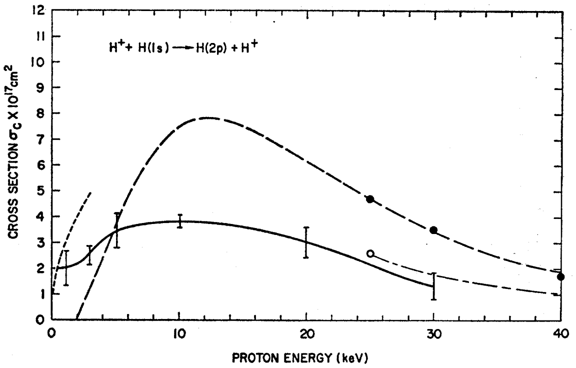

FIG. 3. Cross sections for electron capture into the $2p$ state in H^+-H(1s) collisions. The experimental results are shown as the solid line. The error bars shown at a number of energies embrace 50% of the data points. Also shown are the calculations of Bates and Dalgarno (long dashes) and Jackson and Schiff (filled circles), using the first Born approximation. The cross section at 25 keV calculated by McElroy with allowance for distortion and momentum transfer is shown by the open circle, and a reasonable interpolation of his data is shown by the long and short dashes. One-half the cross section calculated by Bates and Williams for collisions leaving the quasimolecular system formed by the colliding pair in the $2p\pi$ state is shown by the short dashes.

Plotted in Fig. 3 is the cross section for the capture process, together with the available theoretical results. Measurements were made at intervals ranging from 1 keV at the lower energies to 2 keV in the high-energy range. Error bars are given only at energies where sufficient data are available to give meaningful probable errors. The work of Bates and Dalgarno,[12] and of Jackson and Schiff[13] uses the Born approximation. Bates and Dalgarno took a simple interaction potential with only a nuclear-electron term while Jackson and Schiff included a nuclear-nuclear term. It is seen that at energies above about 6 keV, the observed energy variation of the cross section is quite faithfully reproduced by the calculations. More recent theoretical work by McElroy,[14] with allowance for the effects of distortion and momentum transfer, gives values for the cross section at a number of energies within the range 25 to 800 keV. Excellent agreement is observed at 25 keV, the only energy common to experiment and theory. Below 5 keV, the experimental values exceed those predicted by the Born approximation and remain surprisingly large down to about 600 eV. A recent paper by Bates and Williams[15] appears to provide the explanation. They consider slow encounters between H(1s) and H^+, taking into account the coupling between the $2p\sigma$ and the $2p\pi$ states of the quasi-H_2^+ molecule. The degeneracy of these states in the united atom limit was not taken into account in earlier work in which states other than $1s\sigma$ and $2p\sigma$ were ignored. It is found that this coupling

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[11] J. W. R. Fennema, in *Ionization Phenomena in Gases*, edited by P. Hubert and E. Crémieu-Alcan (S.E.R.M.A., Paris, 1964), Vol. I, p. 131.

[12] D. R. Bates and A. Dalgarno, Proc. Phys. Soc. (London) **A66**, 972 (1953).

[13] J. D. Jackson and H. Schiff, Phys. Rev. **89**, 359 (1953).

[14] M. B. McElroy, Proc. Roy. Soc. (London) **272**, 542 (1963).

[15] D. R. Bates and D. A. Williams, Proc. Phys. Soc. (London) **83**, 425 (1964).

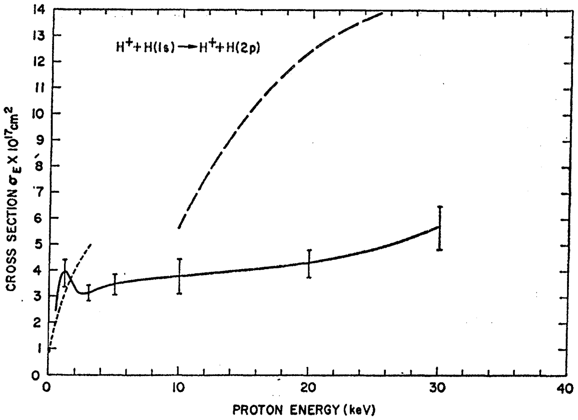

FIG. 4. Cross sections for $1s$-$2p$ excitation of H atoms by proton impact. The experimental results are shown as the solid line. The error bars shown at a number of energies embrace 50% of the data points. The calculations of Bell and Skinner are shown by the long dashed line, and of Bates and Williams by the short dashed line.

is particularly strong and, even in very slow collisions, the probability of the system formed by the colliding pair's being left in the $2p\pi$ state is large. As a result, either the projectile or the target is left in the $2p_{\pm 1}$ states according to whether or not the electron is captured. As the impact energy is increased, transitions to the $2p_0$ state grow in relative importance. At impact energies less than a few keV, Bates and Williams suggested that the cross section associated with the projectile's being left excited is almost the same as that associated with the target's being left excited. Plotted in Fig. 3, therefore, is one-half of the cross section calculated by Bates and Williams for encounters leaving the quasimolecular system in the $2p\pi$ state.

The similarity between the experimental data presented here and the results of Pretzer *et al.*[16] for Lyman-alpha production in proton rare-gas atom collisions is quite remarkable. These authors, too, observe an enhancement of the cross sections at low energy which, in the case of Ne, Ar, Kr, and Xe targets, is even more pronounced than that in the present work. It appears likely that these effects arise from pseudocrossing of potential energy surfaces.

The experimental data for excitation of H($1s$) to the $2p$ state by H^+ impact are shown in Fig. 4 together with the results of recent theoretical work. Considerable theoretical attention has been directed toward the excitation process. First Born-approximation calculations by Bates[17] have been followed by the use of higher approximations. The results of Bell and Skinner[18] (Fig. 4), who took full account of distortion, rotation coupling and back coupling, do not differ greatly from earlier results of Bates, who used the distortion approximation, and Skinner,[19] who also took $2p_0 - 2p_{\pm 1}$ coupling into account. It is seen that at the higher energies, the calculated cross sections exceed those found experimentally by roughly a factor of two although, at the lower energies, agreement with the work of Bates and Williams is again satisfactory.

Inasmuch as the H^+-H data are normalized by comparison with e^--H data, which in turn were normalized to Born-approximation calculations at high energy, it is possible that the cumulative error in normalization may be as high as 30%. The over-all uncertainty in the measured cross sections is estimated to be ±50%.

The measured capture and excitation cross sections are not as similar, below 3 keV, as would be expected on the basis of Bates and Williams' calculations. It should be borne in mind however that the capture cross section is determined by a subtractive procedure and the relative magnitudes of the two cross sections are therefore particularly sensitive to error in the excitation cross section. Inasmuch as the effects of polarization and Doppler shift are subject to uncertainty, too much significance should not be given to the apparent difference in the excitation and charge transfer cross sections at the lowest energies.

CONCLUSION

At energies below about 3 keV the measured excitation and capture cross sections are considerably larger than would be expected through consideration of the adiabatic theory.[20] However, at these energies, satisfactory agreement is observed with the computations of Bates and Willaims[15] who take into account the coupling between the $2p\pi$ and $2p\sigma$ states of the quasi-H_2^+ ion. At higher energies, the earlier calculations appear to overestimate the capture cross section by a factor of 2 although recent computations,[14] in which account is taken of distortion and momentum transfer, remove much of this discrepancy. For the excitation process, the calculated cross sections exceed those determined experimentally by roughly a factor of two at energies above 10 keV. Allowance for cascade would increase the difference between the theoretical and experimental cross section values although this effect is not expected to be large.

The results of the present experiment have application to plasmas of laboratory and astrophysical interest. More significantly, they provide the opportunity for comparison between theoretical and experimental investigation of a heavy-particle collision, involving only one electron, in which the states of all interacting particles are known.

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[16] D. Pretzer, B. Van Zyl, and R. Geballe, Phys. Rev. Letters **10**, 340 (1963).

[17] D. R. Bates, Proc. Phys. Soc. (London) **77**, 59 (1961).

[18] R. J. Bell and B. G. Skinner, Proc. Phys. Soc. (London) **80**, 404 (1962).

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[20] H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Oxford University Press, New York, 1952), p. 441.