

Polarization of Lyman-Alpha Radiation from H^+ - and D^+ -Rare-Gas Charge-Transfer Collisions*†

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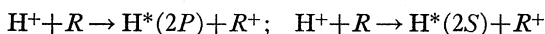
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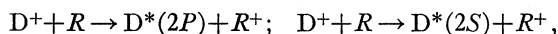
An experimental determination of the polarization of Lyman- α radiation arising from charge-transfer collisions involving energetic protons and deuterons incident on rare-gas atoms has been made in an energy range from 0.5 to 15 keV proton energy. The polarization was computed from the angular distribution of the emitted radiation with respect to the incident projectile direction. Corrections are made to earlier determinations of the product cross sections for the collisions. The cross sections are resolved into contributions from magnetic substates, and the qualitative behavior of these contributions is discussed.

I. INTRODUCTION

EXCITED-STATE charge-transfer cross sections for the reactions



and



where the target atoms R are those of the rare gases have been measured previously in this laboratory, assuming an isotropic spatial distribution of the resulting Lyman- α radiation.^{1,2} Others have recently reported measurements that are in substantial agreement with ours.³⁻⁵ Although an impact formulation of charge-transfer collisions gives a nonzero value for the polarization of the $2P_{3/2} \rightarrow 1S_{1/2}$ radiation, we felt that the cross-section correction due to a finite polarization would have no important influence on the character and magnitude of the results.^{2,6}

Certain observed features of these excited-state cross sections are not completely understood³ although calculations are beginning to close in on the problem.^{7,8}

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† Some of the content of this paper was presented at the Fifth International Conference on the Physics of Electronic and Atomic Collisions, Leningrad, July 1967 (unpublished).

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¹ D. D. Pretzer, doctoral dissertation, University of Washington, 1963 (unpublished); Donavon Pretzer, Bert Van Zyl, and Ronald Geballe, *Phys. Rev. Letters* **10**, 340 (1963). See also the paper by these authors in *Atomic Collision Processes*, edited by M. R. C. McDowell (North-Holland Publishing Co., Amsterdam, 1964).

² D. H. Jaecks, doctoral dissertation, University of Washington, 1964 (unpublished); D. H. Jaecks, B. Van Zyl, and R. Geballe, *Phys. Rev.* **137**, A340 (1965).

³ E. P. Andreev, V. A. Ankudinov, and S. V. Bobashev, *Zh. Eksperim. i Teor. Fiz.* **50**, 565 (1966) [English transl.: *Soviet Phys.—JETP* **23**, 375 (1966)].

⁴ V. Dose, *Helv. Acta Phys.* **39**, 683 (1966).

⁵ G. Ryding, A. B. Wittkower, and H. B. Gilbody, *Proc. Phys. Soc. (London)* **89**, 547 (1966).

⁶ E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, London, 1963), pp. 387.

⁷ L. Wilets and D. F. Gallaher, *Phys. Rev.* **147**, 13 (1966); also see D. F. Gallaher, doctoral dissertation, University of Washington, 1967 for an improved calculation (unpublished).

⁸ I. A. Poluektov and L. P. Presnyakov, in *Proceedings Fifth International Conference on the Physics of Electronic and Atomic Collisions*, Leningrad, 1967, p. 71 (unpublished).

Measurement of any polarization that might exist could reveal in greater detail the nature of the underlying mechanisms. The present experiment was undertaken principally for this reason but also to find the magnitude of any corrections needed for the published cross sections.

Historically, interest in the measurement of the polarization of atomic line radiation began with Skinner and Appleyard and their investigation of electron excitation of mercury vapor.⁹ More recent measurements of the polarization of electron- and proton-induced radiation have given reasonable agreement with existing theories.^{10,11} However, only a limited amount of experimental¹² and theoretical⁷ investigation of the polarization of radiation from excited-state charge-transfer collisions has been carried out.

For this experiment, the usual definition of radiation polarization is assumed, that is,

$$P = \frac{I_{11} - I_{\perp}}{I_{11} + I_{\perp}} \Big|_{90^\circ},$$

where I_{11} and I_{\perp} are radiation intensities at 90° to the incident projectile axis with electric vectors, respectively, parallel and perpendicular to the plane defined by the incident projectile beam axis and the propagation vector of the resulting photon. From the photon electric vector amplitudes an expression is obtained for the radiation intensity observed at some angle θ relative to the projectile axis in terms of the total emitted intensity and the polarization of the radiation. Assuming cylindrical symmetry about the z or projectile axis (see Fig. 1), a familiar analysis¹³ yields

$$I(\theta) = \frac{3I_0}{4\pi} \left[\frac{1 - P \cos^2\theta}{3 - P} \right], \quad (1)$$

where I_0 is the total emitted radiation intensity.

⁹ H. W. B. Skinner and E. T. S. Appleyard, *Proc. Roy. Soc. (London)* **A117**, 224 (1927).

¹⁰ R. H. McFarland, *Phys. Rev.* **133**, A986 (1964).

¹¹ J. Van Eck, F. J. de Heer, and J. Kistemaker, *Physica* **30**, 1171 (1964).

¹² F. J. de Heer, L. Wolterbeek Muller, and R. Geballe, *Physica* **31**, 1745 (1965).

¹³ J. A. Smit, *Physica* **2**, 104 (1935).

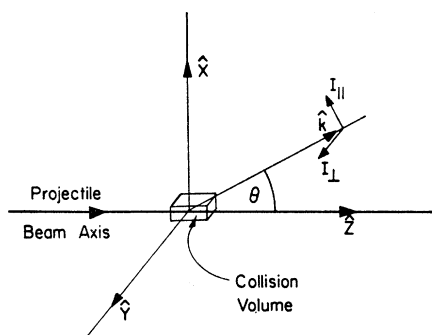


FIG. 1. Collision geometry.

If we consider the collision as populating the three magnetic substates of the $2P$ level, that is, states having magnetic quantum numbers $m_l = \pm 1$ and $m_l = 0$, with quantization axis along the z direction both before and after the collision, we obtain an alternative expression for the radiation polarization in accordance with the following discussion.

For collision times (τ_{coll}) small relative to the spin-orbit precession time (τ_{so}) of the resulting excited atom, the spin-orbit coupling perturbation of the oriented atom may be ignored in the consideration of the excitation processes leading to the population of the various magnetic substates. However, for τ_{so} of the same order or larger than the radiative lifetime of the excited atom (τ_{rad}), full use must be made of the spin-orbit perturbation effects upon the Hamiltonian of the excited atom. In addition, hyperfine effects for certain atoms must be included to obtain proper spin dependence of the emitted radiation polarization. Conservation of angular momentum of the excited atom implies that the spontaneous decay of the $2P_{\pm 1}$ states to the $1S$ state gives rise to photons with electric vectors perpendicular to the z axis, whereas decay of the $2P_0$ state to the $1S$ state yields photons with electric vectors parallel to the z axis. The corresponding intensities are indicated by I_{\perp} and I_{\parallel} , respectively. Denoting the (equal) excitation cross sections of the states $2P_{+1}$ and $2P_{-1}$ by $\sigma(1)$ and that of $2P_0$ by $\sigma(0)$, Percival and Seaton¹⁴ obtain

$$P = [\sigma(0) - \sigma(1)] / [a\sigma(0) + b\sigma(1)], \quad (2)$$

where $a = 2.375$ and $b = 3.749$. From this relation and the definition

$$\sigma_{Total}(2P) = \sigma(0) + 2[\sigma(1)] \quad (3)$$

we can solve for the individual values of $\sigma(0)$ and $\sigma(1)$ in terms of the measured values of P and $\sigma_{Total}(2P)$.

II. TECHNIQUE

A variable energy, mass analyzed, ribbon-shaped beam of positive ions with energy ranging from 0.6 to

¹⁴ I. C. Percival and M. J. Seaton, Phil. Trans. Roy. Soc. London A251, 113 (1958).

15.0 keV was allowed to pass through an entrance-slit system into a collision region containing the target gas under investigation. Typical ion beam currents ranged from 2×10^{-9} A at lowest energies to 1×10^{-8} A at higher energies. Collimation of this ion beam within the collision region was monitored by noting the residual ion beam current impinging on a system of slits, equal in height to the entrance-slit system, placed immediately in front of a Faraday cup collector. Throughout the experiment, current to this system of back slits was never allowed to exceed 5% of the total collected ion beam current. The source of the positive ion beam as well as the beam handling system has been previously described and was used without modification.¹⁵ Commercially available tank gases, trapped by liquid nitrogen, were used as targets.

Differential pumping of the collision chamber was accomplished through the entrance-slit-system aperture across which differential pressure ratios of up to 500:1 were maintained. Ambient background pressures external to the collision chamber were maintained below 5×10^{-6} Torr through the use of a 4-in. oil diffusion pump trapped by a refrigerated baffle system. The actual collision region of projectile beam and target gas was defined by a system of optical collimating slits located in front of a helium and iodine vapor-filled Lyman- α photon detector with a lithium-fluoride window.^{1,15,16} The detector and collimator assembly were attached, as shown in Fig. 2, to the movable curved top of the collision chamber which was fitted with gas seals to confine the outflow of target gas exclusively through the entrance-slit-system aperture. With a high-vacuum rotary motion feedthrough it was possible to vary the relative angle between incident projectile beam axis and the direction of optical observation over a range greater than 55° .

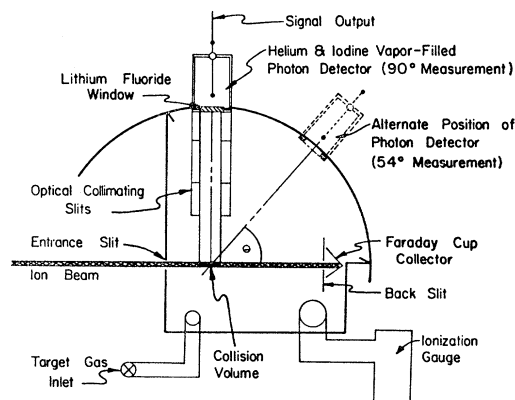


FIG. 2. Collision chamber and photon detector.

¹⁵ G. H. Dunn, doctoral dissertation, University of Washington, 1962 (unpublished); G. H. Dunn, R. Geballe, and D. D. Pretzer, Phys. Rev. 128, 2200 (1962).

¹⁶ R. T. Brackmann, W. L. Fite, and K. E. Hagen, Rev. Sci. Instr. 29, 125 (1958).

All interior surfaces of the collision chamber as well as the interior of the optical collimator were coated with black gold to reduce reflection of the Lyman- α photons and to eliminate radiation resulting from charged particle-surface collisions.^{17,18}

The helium and iodine vapor-filled radiation detector used in this experiment differed substantially from those used previously. It was an integrally sealed device completely contained within the outer vacuum wall of the apparatus. Since the response time of such a device is determined largely by transit times of the positive iodine molecular ion from the region of formation within the detector to the wall of the device, we used a smaller counter in order to increase the maximum counting rate. In practice count rates were always less than 150 counts per second. The correction for lost counts due to the finite response time did not exceed 5% of the largest signals. As previously reported, these photon detectors are sensitive to wavelengths from 1050 Å to approximately 1300 Å.¹⁶

The observations reported here consist of emission radiation from the $2P$ state of the H atom. This state will, of course, be populated by cascade from higher-lying ones. Thus to deduce the cross section for capture into the $2P$ states one must make suitable corrections for lifetimes and cascade effects to the emission cross sections. These corrections are negligibly small for the case of the hydrogen atom in the apparatus used here.

The radiation polarization was computed from the ratio of intensities measured at angles of 90° and 54.5° utilizing Eq. (1). To eliminate effects of the interaction of the ion beam with the residual gas in the collision region, signals measured at a fixed observation angle with the collision chamber evacuated were recorded and subtracted from the signals measured with the target gas in the chamber. Since the detection efficiency of the radiation detector displayed continuous temporal variation, we found it necessary to make repeated measurements at a fixed projectile energy. Signals for alternate angle settings were recorded until sufficient data were accumulated to allow at least five determinations of the polarization at which time the projectile energy was changed to a new value. Target gases and projectiles also were alternated in the course of the experiment and although some deviation of the data from measurement to measurement was observed, no systematic trends were encountered. When changing from proton to deuteron projectiles, adequate precautions were taken to ensure a pure D^+ signal, uncontaminated by residual H_2^+ from the ion source.

Because of the Doppler shift of the wavelength of the radiation from the moving atom and the necessity for intensity measurement at angles other than 90° to the projectile beam axis, it was necessary to eliminate the oxygen wavelength filtering of the radiation detector

used during previous experiments. The loss of wavelength discrimination meant that the target species had to be confined to those having no excited atomic spectral lines within the sensitive region of the detector. The excited argon lines at 1048.2 and 1066.7 Å are close to the cutoff wavelength of the lithium-fluoride window of the detector and do not seriously affect the data (see Sec. III).

During the experiment the current and pressure dependence of the observed radiation signals were measured at each observation angle for each projectile-target gas combination at selected projectile velocities and exhibited the expected linear behavior. At higher projectile velocities, necessitating larger collision chamber electrical potentials, the possibility that spurious signals were induced by electrical breakdown was investigated with negative results. Corrections were made to compensate for the change in the collision volume as a function of the angle of observation. Since the computation of polarization involves only the ratios of observed radiation intensities, absolute calibration of these intensity signals was unnecessary during this experiment. Other electronic signal-processing techniques and data accumulation methods were substantially the same as those used in previous experiments in this laboratory.

III. RESULTS AND DISCUSSION

Figures 3-6 display the radiation polarization for the indicated reactions. Proton and deuteron projectile data are plotted as a function of projectile velocity and energy. Deuteron energies have been scaled by a factor of $\frac{1}{2}$ to allow direct comparison with the proton data. Vertical error bars represent the observed root-mean-square deviation of all of the data points. At selected projectile velocities we indicate by horizontal flags the deviations expected from random fluctuations in the observed photon signals. In most cases the observed rms deviations differ at most by a factor of two from the expected deviations, indicating no serious instrumental errors. The largest deviations of the polarization data occur at the lowest projectile velocities. At these velocities the reaction cross sections as well as the ion beam densities are lowest, necessitating long observation times to obtain a large ($\sim 10^4$) number of events. Changes in detector sensitivity, ion beam current and target gas pressures, although small, have a considerable effect on the scatter of the low-velocity data.

Figures 7-9 display total cross sections for capture into the $2P$ state after correction for polarization for the three target gases investigated. These curves supersede those of Ref. 1 for the targets involved. The most significant changes occur at lowest projectile velocities and in no case do substantial corrections exist in the regions of the structure in the cross sections. In earlier discussions of the $2P$ state cross sections,^{1,2} especially for neon and argon targets, it has been

¹⁷ L. Harris and J. K. Beasley, *J. Opt. Soc. Am.*, **42**, 135 (1952).

¹⁸ J. W. McGowan, *Rev. Sci. Instr.* **38**, 285 (1967).

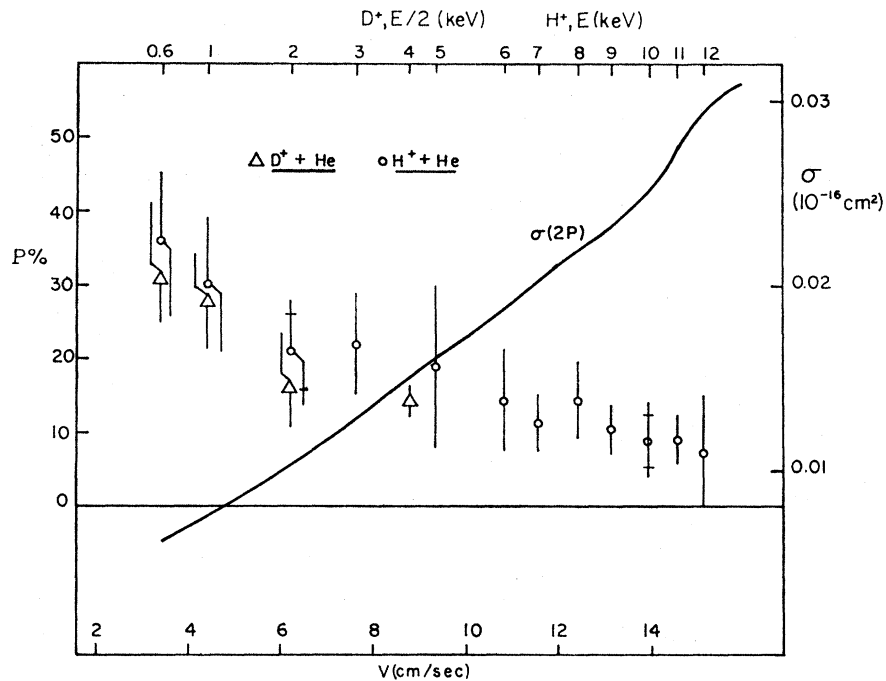


FIG. 3. Lyman- α radiation polarization for proton (and deuteron)-helium collisions. $\sigma(2P)$ is the cross section for Lyman- α production (Ref. 1).

convenient to denote low- and high-velocity maxima which occur in these reactions. It has been suggested that these maxima occur through different coupling processes during the collision. Helium as a target shows no evidence for this complication since it displays only a single maximum in the cross section within the energy range of 1 to 70 keV.

When the polarization fraction is compared with the corrected $2P$ state cross sections, certain features

become apparent which are best observed by resolving the cross sections into the magnetic substate populations, $\sigma(0)$ and $\sigma(1)$. Figures 10-13 show the resolved cross sections for the four reactions $H^+ + He$, $H^+ + Ne$, $D^+ + Ne$, and $H^+ + Ar$.

Considering all reactions, the radiation polarization tends generally to decrease with increasing velocity, but with prominent structure in the cases of Ne and Ar targets. No negative values of the radiation polari-

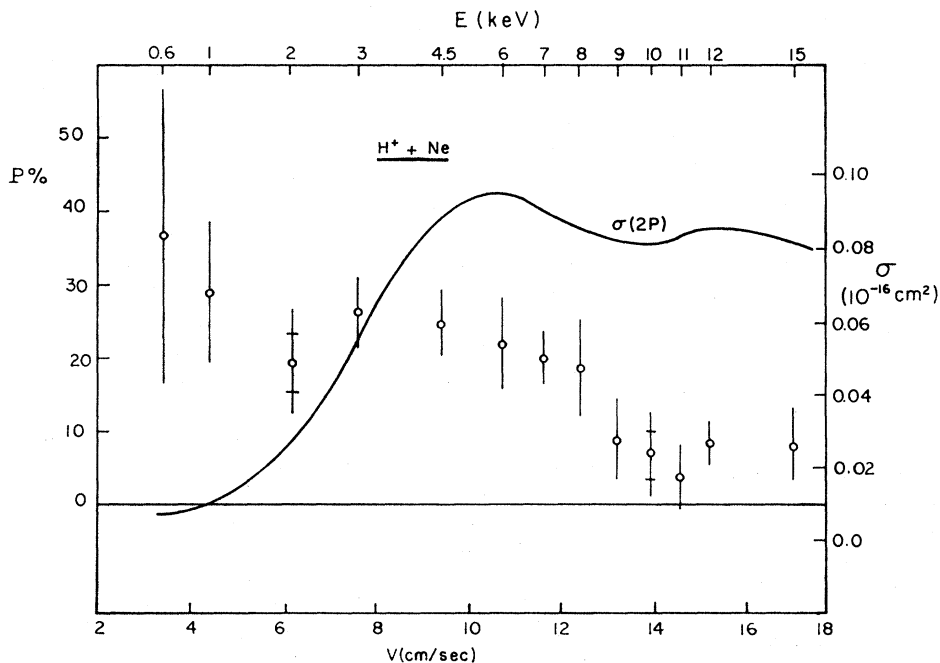


FIG. 4. Same as Fig. 3 except proton-neon collisions.

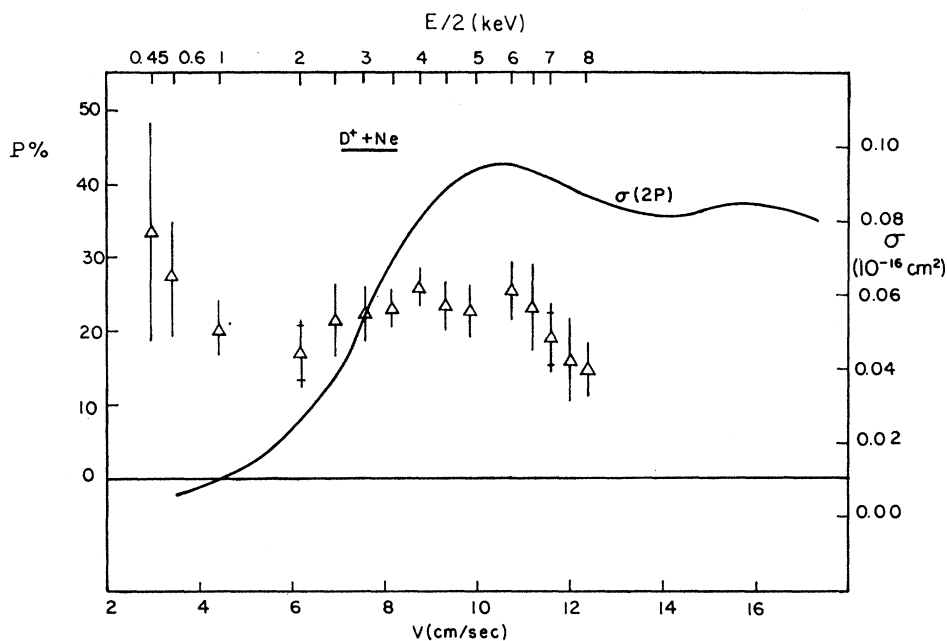


FIG. 5. Same as Fig. 3 except deuteron-neon collisions.

zation were observed other than statistical fluctuations in the data at higher projectile velocity. (See Appendix.)

These results indicate a behavior qualitatively like target atom-projectile distortion during the collision. Such distortion is described by some authors¹⁹ as target atom "polarization," but this terminology is confusing in the present context. It is reasonable to believe that distortion (and electron transfer) occurs preferentially along the internuclear axis, the axis along which the $2P_0$ electron wave function extends. At the lowest projectile velocities the target atom electron distri-

bution has ample time to adjust to the influence of the passing projectile; thus distortion should be largest. Therefore the ratio $\sigma(0)/\sigma(2P)$ should be largest in this energy region. With increasing projectile velocity, the distortion of the projectile-target atom system is reduced because of the reduced target atom electron velocities compared to the projectile velocities. Therefore at higher projectile velocities it is the overlap of the unperturbed wave functions of the target and projectile that determines charge-transfer probabilities and with this overlapping is expected a statistical or equal population of $\sigma(0)$ and $\sigma(1)$. The experimental

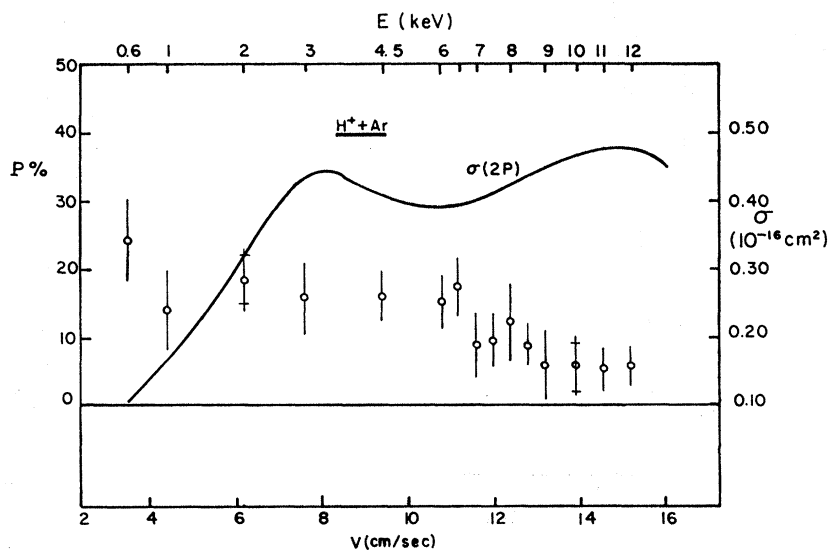


FIG. 6. Same as Fig. 3 except proton-argon collisions.

¹⁹ F. J. de Heer, in *Advances in Atomic and Molecular Physics*, edited by D. R. Bates and I. Estermann (Academic Press Inc., New York, 1966), Vol. 2, p. 357.

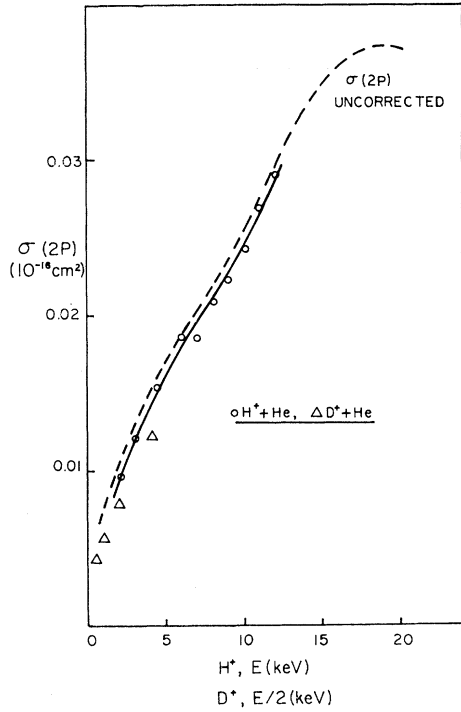


FIG. 7. Cross section for Lyman- α production in proton (and deuteron)-helium collisions corrected for radiation polarization effects. Also shown (dashed curve) is the uncorrected cross section as reported in Ref. 1.

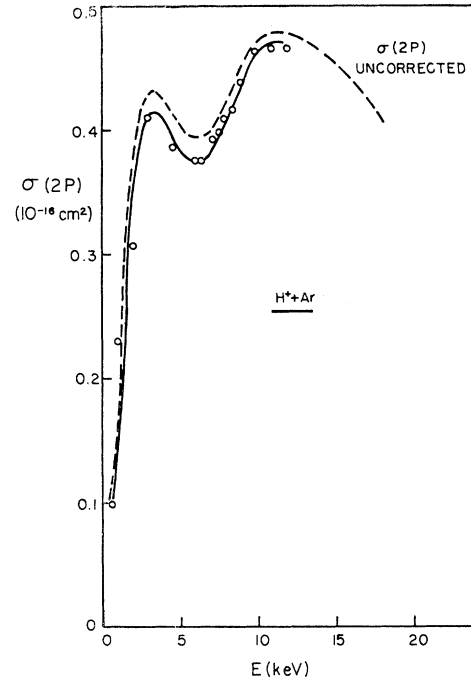


FIG. 9. Same as Fig. 7 except for proton-argon collisions.

results illustrate the behavior suggested by these considerations.

Independent experimental substantiation of this view of the distorted target atom-projectile system is found

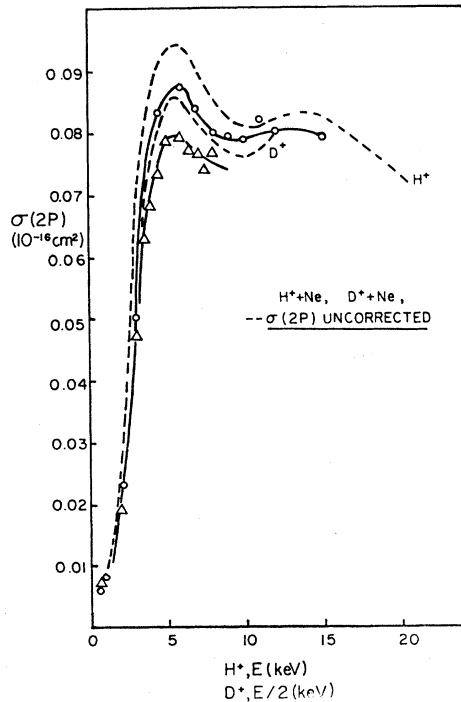


FIG. 8. Same as Fig. 7 except for proton (and deuteron)-neon collisions.

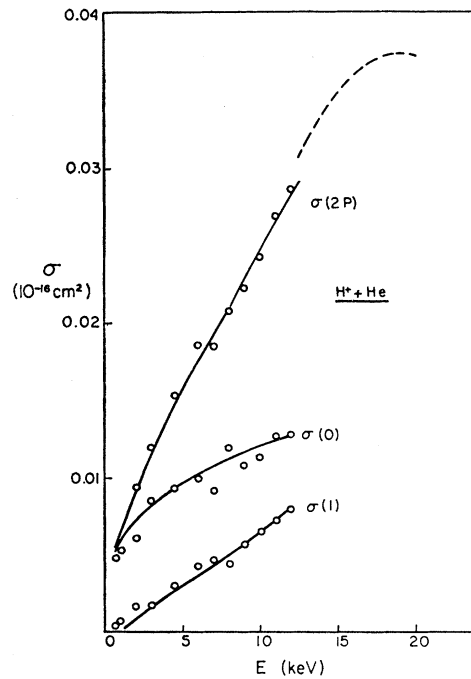


FIG. 10. Corrected $2P$ state cross section for proton-helium collisions showing the magnetic substate populations deduced from Eq. (2). $\sigma(0)$ is for $m_l=0$, $\sigma(1)$ is for $m_l=\pm 1$.

in the investigation of target excitation collisions between protons and helium atoms compared to similar collisions between fast neutral hydrogen atoms and helium atoms.¹¹ The former case should exhibit more distortion with associated increase in the magnitude of the radiation polarization. Over the projectile energy range investigated by Van Eck *et al.*,¹¹ the radiation polarization for proton impact was larger than that for hydrogen-atom impact.

The similarity of the radiation polarization resulting from proton and deuteron collisions with a given target gas, a similarity found in all of the present work as well as past excited state cross-section measurements, can be explained if the interaction involves only the electronic structure of target and projectile. The initial excitation phase should yield populations of the sub-

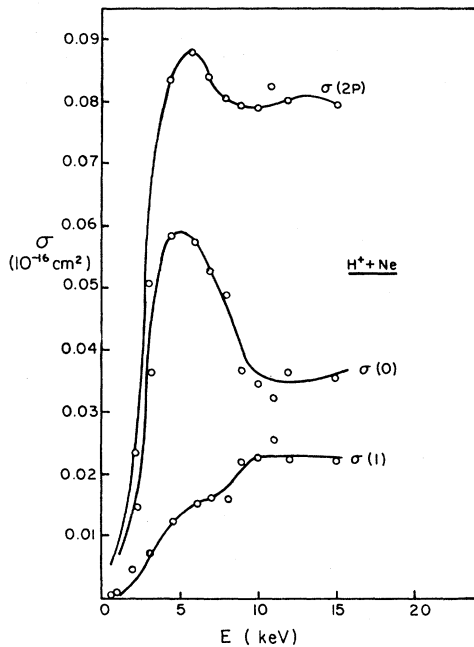


FIG. 11. Same as Fig. 10 except for proton-neon collisions.

levels for hydrogen and deuterium atoms which are the same. In the radiative phase, however, a more detailed analysis must be made following that of Percival and Seaton¹⁴ for the hydrogen atom which gives the result in Eq. 2. We have carried out a treatment of the same kind for the deuterium atom, with the result that the following expression for D-atom Lyman- α polarization is obtained:

$$P = \frac{\sigma(0) - \sigma(1)}{a'\sigma(0) + b'\sigma(1)},$$

where $a' = 2.341$ and $b' = 3.681$. It is seen that these coefficients differ only slightly from those for hydrogen. Thus the spin-dependent (hyperfine) treatment of Percival and Seaton, rather than the earlier work by

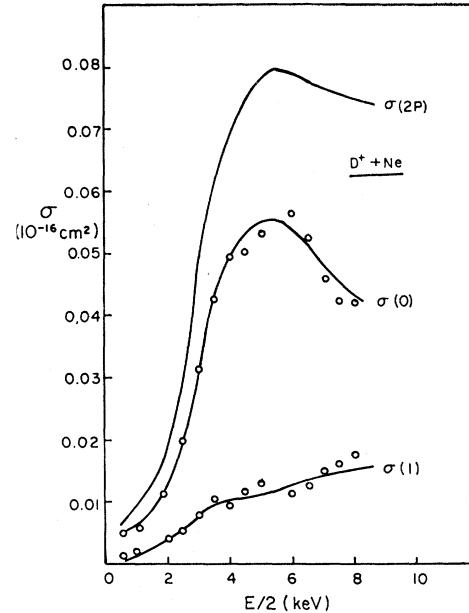


FIG. 12. Same as Fig. 10 except for deuteron-neon collisions.

Oppenheimer and Penney,²⁰ seems best to fit the experimental results.

Theoretically, therefore, one concludes that $H^*(2P)$ and $D^*(2P)$ formed by charge transfer (or direct excitation) should produce similar radiation patterns. This conclusion, at least for charge transfer, is borne out by our experimental results.

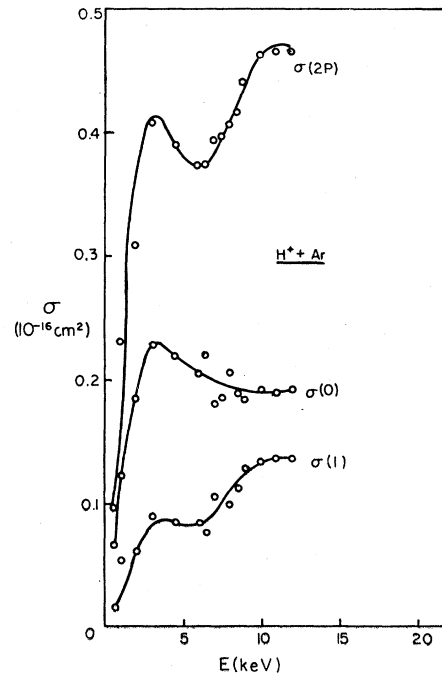


FIG. 13. Same as Fig. 10 except for proton-argon collisions.

²⁰ J. R. Oppenheimer, *Phys. Rev.* **32**, 361 (1928); W. G. Penney, *Proc. Natl. Acad. Sci.* **18**, 231 (1932).

Each of the three observed reactions is now considered in detail.

A. $H^+ + He$

As mentioned previously, this target gas is the only one of the rare gases investigated which does not exhibit two maxima in its cross section for Lyman- α emission (Fig. 10). Independent investigations of this reaction have been made at overlapping and greater projectile velocities with no evidence for another maximum in the $2P$ and $2S$ state cross sections.³⁻⁵ The radiation polarization for this reaction exhibits no noticeable structure beyond fluctuations and $\sigma(0)$ is everywhere larger than $\sigma(1)$.

B. $H^+ + Ne, D^+ + Ne$

The available projectile velocity for this reaction (Figs. 11 and 12) covers the range through both maxima in the $2P$ state cross section. The marked decrease in radiation polarization at projectile velocities of $\sim 12 \times 10^7$ cm/sec appears well correlated with the decrease in the $2P$ state cross section on the high-velocity side of the low-velocity maximum.

Several mechanisms for the depolarization of impact radiation or disorientation of the excited atom are known, including cascade contributions from higher excited states, stray transverse magnetic fields in the collision region and multiple collisions of the excited atoms. Linearity of the signal with target gas pressure seems to obviate multiple collisions and a measurement of stray magnetic field within the collision chamber indicated fields of less than 10 G. Applying the analysis of Skinner,²¹ the following expression is derived for the influence of magnetic fields on the polarization of the radiation:

$$[2\pi\tau\omega]^2 = [P_0/P]^2 - 1,$$

where, P_0 is the polarization at zero magnetic field, P is the polarization with transverse magnetic field, τ is the lifetime of the excited level, and ω is the Larmor frequency $eB/4\pi m_e$ for the transverse component of the magnetic field. For a 10-G transverse field, a change in the radiation polarization of only 4% is expected. Earlier estimates² of the cascade contributions to the $2P$ state cross sections showed negligible contribution from higher-lying excited states. Energy-dependent focusing of the positive-ion beam within the collision region was investigated and eliminated as a cause of apparent polarization.

Thus the origin of the decrease in the radiation polarization at this particular velocity cannot be attributed to these secondary effects.

If, as previously suggested,² two different mechanisms of electron capture into the $2P$ excited state are

responsible for appearance of the dual maxima in the cross sections, it is reasonable to expect a change with energy in the magnitude of the radiation polarization. A comparison of the $2P$ and $2S$ excited state cross sections with that for total charge transfer in several cases² led to the conjecture that the low-velocity maximum occurs through an intermediate coupling to the ground state of the resulting hydrogen atom. In addition ground-state coupling should involve the $m_l=0$ magnetic substate most strongly. The large contribution of $\sigma(0)$ to the low-velocity maximum in $\sigma(2P)$ as illustrated by Figs. 11 and 12 is consistent with this suggestion.

C. $H^+ + Ar$

Again the available velocity of the projectile covers the region (Fig. 13) of both maxima in the $2P$ cross section. Although exhibiting substantial scatter, the radiation polarization decreases in magnitude at projectile velocities greater than $\sim 12 \times 10^7$ cm/sec. Here, correlation with the decrease in the $2P$ state cross section on the high-velocity side of the low-velocity maximum is less apparent than with the case of neon, but scatter of the data as well as a different gross structure of the cross-section maxima may obscure the correlation.

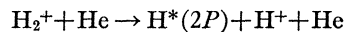
A lower over-all magnitude of the radiation polarization for this reaction is observed. This effect may be due to the presence of spectral lines of excited argon at approximately 1050 Å which are unrelated in orientation to the excited hydrogen atom radiation. No data were taken for the $D^+ + Ar$ reaction during the present experiment.

ACKNOWLEDGMENT

We wish to acknowledge the contributions of Mrs. Patricia Huff for operation of the equipment and for help with data reduction. M. Harnois also assisted with data reduction.

APPENDIX

In order to determine the possible existence of residual or instrumental effects that mimic polarization, the angular distribution of Lyman- α radiation from the reaction



was measured. Van Zyl *et al.*²² conclude that the principal source of Lyman- α radiation for this reaction at low projectile energies is the direct breakup of the projectile into an excited H atom and proton. In addition it is probable that this radiation should exhibit an isotropic spatial distribution with respect to the projectile-beam axis. This belief is based upon an

²¹ H. W. B. Skinner, Proc. Roy. Soc. (London) **A112**, 642 (1926).

²² B. Van Zyl, D. H. Jaecks, D. D. Pretzer, and R. Geballe, Phys. Rev. **136A**, 1561 (1964).

assumed random spatial orientation of the proton-hydrogen atom axis with respect to the projectile-beam direction. This new internuclear axis, rather than the projectile-beam axis, acts as the quantization direction for the resultant excited hydrogen atom. While largely conjectural, this hypothesis is substantiated by the measured Lyman- α -radiation polarization and also the measurement of Balmer- α -radiation polarization from

this same reaction.²³ Measurements at 3- and 6-keV projectile energy yielded zero polarization for the Lyman- α radiation with rms deviation of the data consistent with that from the proton projectile reactions. We therefore conclude that no serious instrumental effect influences the polarizations reported here.

²³ D. H. Jaecks and F. J. de Heer (unpublished).

PHYSICAL REVIEW

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Spontaneous Emission in the Presence of a Prescribed Classical Field*

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The effect of the application of a classical driving field on the spectrum of spontaneous emission from a simple physical system is studied. The physical system consists of an ensemble of two-level atoms interacting with a relaxation mechanism. The single Lorentzian line-shape characteristic of the power spectrum of spontaneous emission for the undriven case is split into components by the driving field. This splitting is associated with the establishment of definite phase relations between the corresponding components of the field spectrum.

INTRODUCTION

WE will investigate the spectral distribution of the spontaneous emission from a system of atoms subjected to a strong, near-resonant field. The material system is modeled by an ensemble of two-level atoms interacting with a relaxation mechanism.

The spectral distribution of the spontaneous emission will be unaffected by the driving field if the latter is not sufficiently large to significantly alter the state of the atomic system in a relaxation time.¹ If the driving field is sufficiently large we expect the spectral distribution to be changed from the undriven case. When the frequency associated with the interaction energy (of the atomic currents with the electromagnetic field) is small compared to the resonance frequency, useful calculational techniques are based on treating the interaction Hamiltonian as a perturbation of the full Hamiltonian. This technique is not valid for the present problem. Rautian and Sobelman² have considered the situation that the material system is coupled to many modes of the radiation field, but that initially only one mode of the field is in a high-energy eigenstate. They obtained a solvable finite set of equations for the atom field probability amplitudes by truncating the infinite set to

correspond to a small number of multiphoton processes. It appears to us that the validity of this procedure is limited to relatively small initial field energies. Bergmann³ has investigated the problem of spontaneous emission from a two-level system with incident beams of radiation which are initially either in a coherent state or in an n -photon state. His treatment attempts to avoid a perturbation-theory approach and uses approximations which retain only diagonal elements of the field time-development operator. In addition to these approximations his equations of motion are restricted to material two-level systems which cannot develop into mixed states. We feel it is necessary to consider the effect of mixed states, and indeed the main features of the spontaneously radiated power spectrum for large fields are simply related to the time development of these states.

In our work we take the large driving field to be classical and prescribed in its time dependence. The Hamiltonian now includes separate terms which describe the interactions of the atomic current with the prescribed classical field and with the small quantum-mechanical field which causes spontaneous emission. We compute the spectral distribution of the spontaneous emission to second order in the small interaction. The general solution gives the spectral power radiated in terms of the characteristic frequencies which describe the problem, and the atomic and relaxation-mechanism parameters. This solution has been used as a basis for determining the fundamental noise properties

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† This work was done while the author was at Technical Research Group, Division of Control Data Corporation, Melville, N. Y.

¹ I. R. Senitzky, *Phys. Rev.* **119**, 1807 (1960).

² S. G. Rautian and I. I. Sobelman, *Zh. Eksperim. i Teor. Fiz.* **41**, 456 (1961) [English transl.: *Soviet Phys.—JETP* **14**, 328 (1962)].

³ S. M. Bergmann, *J. Math. Phys.* **8**, 159 (1967).