like ion of nuclear charge Z by converting to the $1/Z$ system of units. The new unit of length is $1/Z$ a.u.; the new unit of energy, $1/Z^2$ a.u.; and the unit of field intensity, $1/Z^3$ a.u.

[13]E. L. Ince, Ordinary Differential Equations (Dover Publications, Inc., New York, 1926).

[14]L. B. Redei, Phys. Letters 1, 191 (1962).

[15]Here we have surpressed the time dependence.

[16]Our method of solution to the ξ equation yields both upper and lower bounds to ΔZ_1 for a given ΔE. After obtaining these bounds to high precision (10 significant figures), we then average them for use in solving the η equation.

[17]All calculations were carried out with double-precision arithmetic. The determination of upper and lower bounds to ΔE, differing by less than 1 part in 10^8 and involving the determination of ΔZ_1 to within 1 part in 10^{10}, takes less than 20 sec on an IBM 7094.

[18]C. A. Coulson, A. Maccoll, and L. E. Sutton, Trans. Faraday Soc. 81, 106 (1952); P. W. Langhoff, J. D. Lyons, and R. P. Hurst, Phys. Rev. 148, 18 (1966).

[19]For the determination of the 2nd order term, see Ref. 9. The exact 4th order term is due to A. Dalgarno, Quantum Theory, edited by Bates, (Academic Press, Inc., New York, 1961) Vol. 1, p. 187.

[20]A. Messiah, Mécanique Quantique (Dunod, Paris, 1965) Vol. I, pp. 194–202.

[21]This form includes the first element of the correction term given by Messiah; Eq. (45), p. 196 of Ref. 20. For the 1s state of hydrogen at a field strength of 0.05 a.u., the phase shift obtained from Eq. (26) has converged to within 1 part in 10^4 at $\eta_o = 40$ a.u. Using Eq. (24), one obtains the same degree of convergence only for $\eta_o > 80$ a.u.

[22]Reference 20, pp. 336–342.

[23]Indeed, for fields greater than 0.12 a.u. the slope of $\delta(\Delta E)$ becomes very nearly constant. It is then impossible to distinguish any resonant behavior at all.

[24]We assume that there exist two real roots. If the roots are imaginary, then b_η is always less than unity and it becomes impossible to establish a partial divergence theorem. We have not encountered this situation.

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Measurement of an Emission Cross Section for the Collision Reaction $H^+ + N_2(X^1\Sigma_g^+, v=0) \rightarrow H(3p,3d) + N_2^+(B^2\Sigma_u^+, v=0)$ Using Photon-Coincidence Techniques*

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The measurement of the cross section for the emission of a Balmer-α and a $N_2^+(0,0)$ first-negative-band photon from a hydrogen atom and a N_2^+ molecule excited simultaneously in the same $H^+ + N_2$ electron-capture collision has been performed in the proton velocity range from 0.54×10^8 cm/sec to 2.40×10^8 cm/sec. The measurement has been accomplished using the technique of single photon-coincidence detection. To within the random error of the experiment, the cross section has only a single-peak structure with a maximum value of 1.2×10^{-18} cm^2 at a velocity of 1.15×10^8 cm/sec. The magnitude and shape of the measured cross section is found to be in good agreement with a cross section calculated by assuming that final-state excitations of the two product systems are uncorrelated.

I. INTRODUCTION

The specification of a particular collision reaction between two systems, both of which have internal structure, must necessarily include a statement of the structural state of both product systems. If the systems are prepared in given initial states and total rather than differential reaction cross sections are desired, the specification of the final internal states of both systems defines a unique collision reaction. However, in previous cross-section measurements for such systems the final states of both colliding partners have not been specified.

An experiment in which the initial and final states of both reacting systems are specified has been carried out at our laboratory for the specific case of simultaneous excitation of the 3p or 3d state of hydrogen and the 0 vibrational level of the $B^2\Sigma_u^+$ state of N_2^+ (hereafter referred to as the "B state") in electron-capture collisions of protons with nitrogen molecules. Specifically, we have measured the cross section for production of Balmer-α radiation from the 3p or 3d states of hydrogen and 3914 Å N_2^+ band emission in a single-proton–nitrogen collision. This measurement has been accomplished by counting single photons in coincidence.

This method for measuring collision-reaction cross sections consists, in brief, in the detection of the two photons resulting from the excitation of

both product systems in a single collision. Both detectors view the same "observation region" and a signal pulse is produced whenever both photons are detected within a time τ.

II. DETERMINATION OF COINCIDENCE DETECTION PROBABILITY

Because atoms and molecules have finite radiative lifetimes, one must take into account those cases in which excitation of the desired states occurs in a single electron-capture collision, but the two emissions occur at times separated by more than the coincidence resolving time, τ. It is also necessary to account for the fast, excited, hydrogen atoms which pass out of the observation region before emitting.

To account for these effects, we define a function P_{3m} as the fraction of the collisions of interest occurring in the observation region for which the emissions from the two excited systems occur at times separated by less than τ and in view. The probability that if a collision takes place in the observation region it will take place in the time interval dt_0 about t_0 is

$$dp_0 = (L/v)^{-1} dt_0 ,$$

where the observation region extends from $z = 0$ to $z = L$ and $t = 0$ is the time of arrival of the proton at $z = 0$. If the excitation takes place at time t_0, the probability that the moving system (the hydrogen atom) emits a photon in dt_M about $t_M \geq t_0$ is

$$dp_M = \tau_M^{-1} \exp[-(t_M - t_0)/\tau_M] dt_M ,$$

where τ_M is the lifetime of the excited state of the moving system. Similarly, the probability that the "stationary" recoil system (the N_2^+ ion) emits a photon in dt_S about $t_S \geq t_0$ is

$$dp_S = \tau_S^{-1} \exp[-(t_S - t_0)/\tau_S] dt_S .$$

The probability P_{3m} is the product $dp_0 dp_M dp_S$ integrated over suitable limits. Since the moving system must emit within the viewing region, the limits on the variable t_M are

$$t_0 \leq t_M \leq L/v .$$

The limits on t_S are then set by the requirement that the time interval between emissions must be less than τ. In the case that $\tau \geq L/v$, the limits are

$$t_0 \leq t_S \leq t_M + \tau .$$

The limits on the variable t_0 are

$$0 \leq t_0 \leq L/v .$$

Hence

$$P_{3m} = \int_{t_0=0}^{L/v} \int_{t_M=t_0}^{L/v} \int_{t_S=t_0}^{t_M+\tau} dp_S dp_M dp_0$$

with the result that

$$P_{3m} = 1 - [\tau_S/(\tau_S + \tau_M)] e^{-\tau/\tau_S} + \frac{v}{L} \left\{ \frac{\tau_S^2 \tau_M}{(\tau_S + \tau_M)^2} \left[1 - \exp\left(-\frac{L}{v} \frac{\tau_S + \tau_M}{\tau_S \tau_M} \right) \right] \times e^{-\tau/\tau_S} - \tau_S [1 - \exp(-L/v)\tau_M] \right\} . \quad (2.1)$$

Notice that P_{3m} involves the variables L and v only as the ratio L/v.

The specialization to the case where $\tau \geq L/v$ results from the fact that the maximum value of L/v used in this experiment is approximately 30 nsec and the value of τ used is 70 nsec. The P_{3m} functions are plotted in Fig. 1 for $m = s$, p, and d as a function of L/v to indicate their shapes and relative values. The values of τ_M are 160, 5.4, and 15.6 nsec, respectively, for $m = s$, p, d,[1] and $\tau_S = 65.8$ nsec.[2]

In the derivation of Eq. (2.1), considerations of jitter in the detection apparatus and variations of detection efficiency with position of the emitter in the viewing region have been neglected.

If we define $\sigma_c(3m, B)$ as the cross section for the

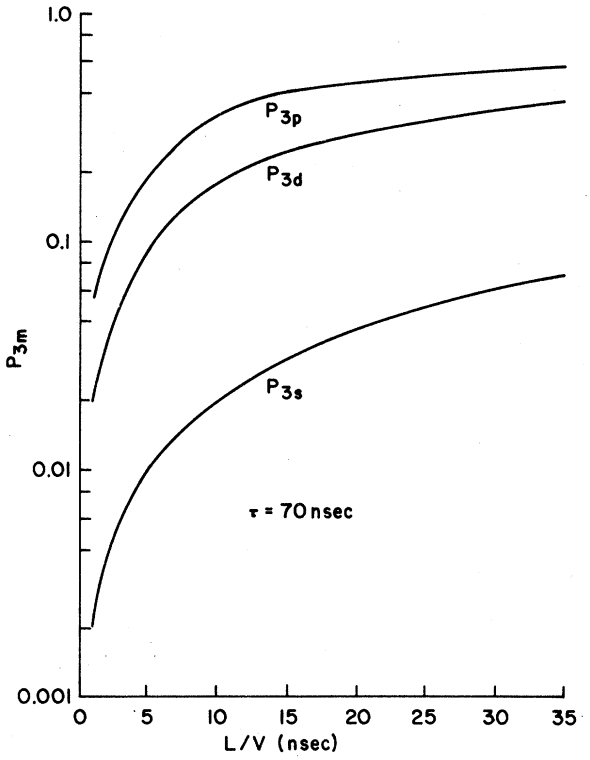

FIG. 1. Coincidence detection probability functions for the $n = 3$ states of hydrogen and the $B^2\Sigma_u^+ (v = 0)$ state of N_2^+ as a function of L/v.

production of a Balmer-alpha photon from a hydrogen atom excited to the $3m$ state and a 3914 band photon from a N_2^+ molecule excited to the B state in the same electron-capture collision, the number of such coincident emissions observed for a specific hydrogen transition ($3s \rightarrow 2p$, $3p \rightarrow 2s$, or $3d \rightarrow 2p$) per unit time is

$$\dot{N}_{3m} = (InL/e)A_1A_2P_{3m}\sigma_c(3m,B) ,$$

where I is the proton current at the point of observation, n is the target gas number density, e is the electronic charge, and A_1 and A_2 are the fractions of the number of photons emitted per unit volume which are detected by the respective detectors. Including all degenerate hydrogen states, the total signal rate is

$$\dot{N}_R = (InL/e)A_1A_2[P_{3s}\sigma_c(3s,B) + P_{3p}\sigma_c(3p,B) + P_{3d}\sigma_c(3d,B)] . \quad (2.2)$$

Figure 1 shows that P_{3s} is significantly smaller than P_{3p} and P_{3d}. This suggests that the $3s$ contribution to the signal may be neglected if $\sigma_c(3s,B)$ is not too large.

If $\sigma_c(3s,B)$ is no larger than the estimate resulting from the assumption that the cross sections, $\sigma_c(3m,B)$, for different m have the same relative sizes as do the corresponding total cross sections for production of Balmer-α emissions,[3] σ_{3m}, then it is possible to determine the proton velocity range for which this contribution may be neglected. Using the σ_{3s} cross section reported by Hughes[4] and the total Balmer-α cross section of Murray *et al.*,[5] it is found that the contribution to the signal from $3s \rightarrow 2p$ transitions, i.e., $P_{3s}\sigma_c(3s,B)$, is less than 10% of the total contribution for proton velocities less than 1.85×10^8 cm/sec and the worst possible value for σ_{3p}/σ_{3d} (i.e., for $\sigma_{3p} = 0$). If $\sigma_{3p} \gg \sigma_{3d}$, the estimated contribution is reduced to a maximum of 4% in this velocity range.

Neglecting the $3s \rightarrow 2p$ and cascade contributions to the signal, Eq. (2.2) becomes

$$\dot{N}_R = (InL/e)A_1A_2[P_{3p}\sigma_c(3p,B) + P_{3d}\sigma_c(3d,B)],$$

$$\text{or } \dot{N}_R = (InL/e)A_1A_2G ,$$

$$\text{where } G \equiv P_{3p}\sigma_c(3p,B) + P_{3d}\sigma_c(3d,B) .$$

The most straightforward method of obtaining values for $\sigma_c(3p,B)$ and $\sigma_c(3d,B)$ would be to measure G for two different values of τ (or L) and solve the resulting two equations. Unfortunately, the variations of P_{3p} and P_{3d} with τ or L lead to degenerate equations in the L/v range of interest. This result indicates that there is little hope of experimentally resolving the measured quantity G into the individual cross sections $\sigma_c(3p,B)$ and $\sigma_c(3d,B)$ in an experiment such as the one reported here.

Another method of obtaining meaningful results from the measured signal is to determine the direct sum of the cross sections $\sigma_c(3p,B)$ and $\sigma_c(3d,B)$ by replacing both P_{3p} and P_{3d} with $P_e = \frac{1}{2}(P_{3p} + P_{3d})$. The maximum possible error in the P_{3m} values introduced by this approximation is $\simeq 40\%$ at the higher velocities and 20% at the lower velocities. Estimates[6] based on knowledge of total cross sections for production of Balmer-α radiation yield an error of 20% at the higher velocities and 10% at the lower velocities. Since this is the order of the accuracy of the experiment, this approximation seems appropriate and is used to obtain our final result.

The appropriate relation between cross section and signal is

$$\dot{N}_R = (InL/e)A_1A_2P_e[\sigma_c(3p,B) + \sigma_c(3d,B)] . \quad (2.3)$$

This expression may also be written in terms of excitation cross sections $\sigma(3m,B)$ by use of the relation $\sigma_c(3m,B) = f_{3m}f_B\sigma(3m,B)$ where the f's are the fractions of the respective excited systems which give the required emissions.

III. PROCEDURE AND RESULTS

A. Apparatus and Experimental Procedure

The ion-beam system used here is described in detail by Murray.[7] Protons are produced in an Oak Ridge type hydrogen rf discharge, extracted, accelerated, focused, magnetically mass analyzed, and finally collimated at the entrance to the collision chamber. The limits of proton beam energy are about 2 and 30 keV, and the beam-energy resolution is about 50 eV at the lower energies and 200 eV at the higher energies.

The collision chamber is shown schematically in Fig. 2. The light emitted from the collision region at a distance of 17.8 cm into the collision chamber is collected by plano-convex lenses of small f number ($f = 0.70$), and the image of this

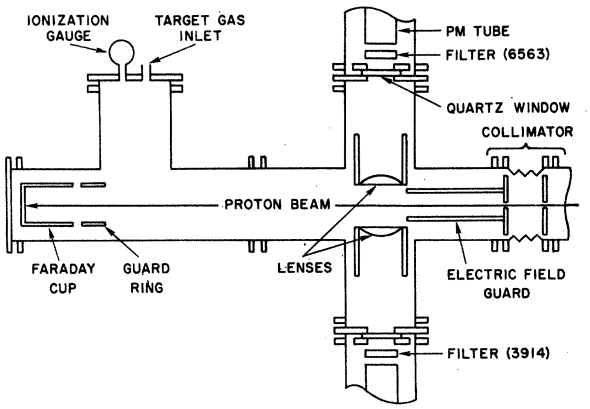

FIG. 2. Collision chamber.

region is focused onto the photocathodes of RCA-7265 PM tubes after passage through quartz vacuum windows and interference filters. The beam diameter at this point is about 0.3 cm. The length of beam viewed is determined by the width of the photocathodes and the effective magnification of the lenses. The two lenses are individually adjusted in position to give the same magnification and to insure that each photometer is looking at the same length of beam and at the same position on the beam. The detection geometry is chosen to maximize the signal-to-noise ratio. (This ratio is derived in Appendix I.) A 1.7-cm-long viewing region was found to be optimum for the lenses used. Since for this geometry, light strikes the interference filters at angles as large as 20°, filters having fairly wide and flat transmission bands are used. This has the effect of keeping the transmission at the wavelengths of interest large and nearly independent of angle of incidence.

The 6563 Å filter has a peak transmission of 80% and a half-width of 200 Å, while the 3914 Å filter has a peak transmission of 30% and a half-width of 113 Å. Spectroscopic scans[8] and measured band ratios[9] verify that, although the filter band passes are rather large, emissions other than Balmer α and N_2^+ 3914 band emissions do not contribute significantly to the signal.

The target gas pressure is monitored with an ionization gauge calibrated against a McLeod gauge. (The geometry is chosen to minimize the Ishii effect.) Signal measurements are made with a target gas pressure low enough ($\simeq 10^{-4}$ Torr) that collisional quenching and second-order processes are negligible. Linearity of signal with beam current and pressure further confirm this.

The signal-detection system consists of two detector channels, a switching module, a coincidence detector, and two scalers (see Fig. 3). The detection channels each consist of a photometer (PM tube plus lens and interference filter), a preamplifier, a double-delay line amplifier (DDL), and a pulse-height analyzer (PHA). Each detection channel is operated as a single-photon detector. The active pulse-shaping components of the detection channels are designed to give output pulses having fast rise times ($\simeq$10 nsec) so that accurate time-correlation measurements between pulses from each channel can be made. The signal-pulse counting rates in the two channels are directly proportional to the total cross sections for the production of the emissions detected.

The switching module performs the following function: The signal from the Balmer-α channel is switched, at a rate of 12.5 times per second, either through or bypassing a fixed delay. Simultaneously, the output of the coincidence detector is switched to one or the other of two fast scalers. The result is that accidental coincidences and accidental plus real coincidences are alternately counted. A simple subtraction of the final tally on the two scalers yields the number of real coincidences. This procedure is followed in order to reduce errors arising from fluctuations and drifts that occur in the beam current, coincidence resolving time, and detector sensitivities over the relatively long integrating times used (1–2 h).

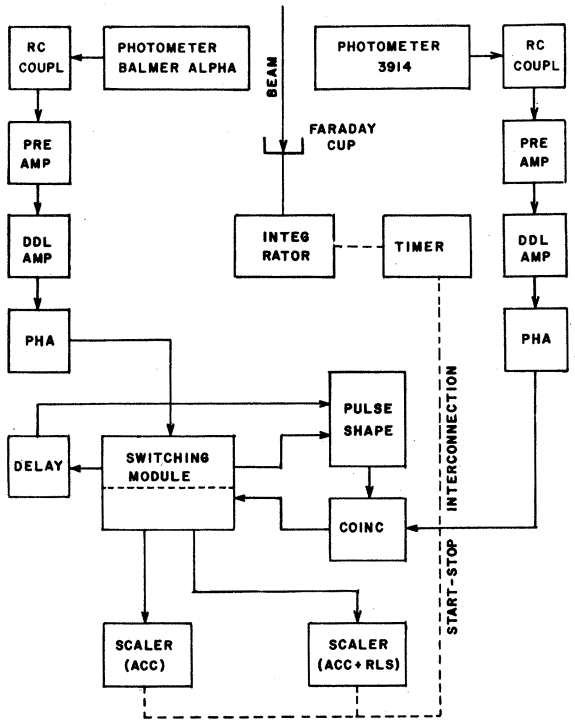

FIG. 3. Signal-detection electronics.

Further details of the detection system and the switching module are presented elsewhere.[6,10]

Several checks are made for possible systematic errors introduced by the detection system. The accuracy of signal recovery is determined by admitting pulses at a rate of 1 pulse per second from a mercury pulser to both detection channels. The resulting coincidence signal is recovered from accidental coincidences with a standard deviation consistent with the number of observed accidental counts. It is verified that when the proton beam is absent or a fixed delay is introduced in one channel, the real coincidence signal is zero. It is also observed that when the photometers are illuminated by independent light sources, no real coincidence signal results.

The system "cable curve"[11] is shown in Fig. 4. The rise and fall times of the cable curve are consistent with the measured 5-nsec "jitter" in the transit-time difference for the two channels. The observed cable curve and jitter do not include transit-time differences between the two photomultipliers. However, variations in transit times for the RCA-7265 PM tubes are typically only about 3 nsec so only a small error can result from this exclusion. The value of τ chosen for the experiment is determined by maximizing the function P_e^2/τ which occurs as a factor in the signal-to-noise ratio (see Appendix I). The resulting optimum value for τ in our experiment is 70 nsec (which is approximately the same as the lifetime of the B state of N_2^+, the longest lived of the two excited states being observed).

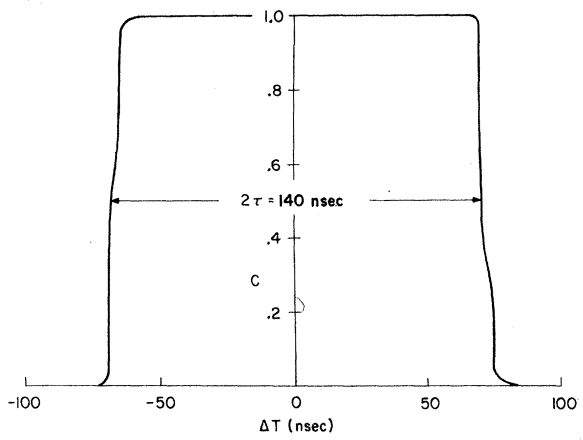

FIG. 4. Cable curve of coincidence detection system (see Footnote 8).

When coincidence signal data are being collected, the scalers and the beam-current integrator are interconnected, thus terminating the signal-integration period when a preset effective beam charge, C, has been collected. An effective integrating period of approximately $\frac{1}{2}$ h is used for each measurement. Because of the method of detection this corresponds to an actual charge $2C$ collected over a time of 1 h. For each set of coincidence signal measurements the target gas pressure, the number of signal counts at each PHA (N_H and N_N), and the corresponding beam charge, C', collected in a time T' are measured. Typical values occurring during one set of measurements at a proton velocity of 1.50×10^8 cm/sec (11.8 keV) are displayed in Table I.

At a particular beam velocity, approximately 10 determinations of N_R are made. The theoretical and experimental standard deviations computed for this data set are always nearly equal. The 70% confidence interval is also computed and is used to represent the random error in $\bar{N}_R$.

B. Data Reduction

The measured value of N_R is related to the emission cross sections by the approximate equation

$$N_R = (CnL/e)A_1A_2P_e[\sigma_c(3p,B) + \sigma_c(3d,B)]\exp(n\sigma_{10}d)\,, \tag{3.1}$$

where C is the effective beam charge collected at the Faraday cup and σ_{10} is the total electron-capture cross section for proton impact on nitrogen. The exponential factor corrects for the loss of protons from the beam between the point of optical observation and the point at which the charge is collected. Similarly, the individual detector counts are

$$N_H = (C'nl/e)A_1Q\exp(n\sigma_{10}d)$$

and

$$N_N = (C'nL/e)A_2\sigma_{3914}\exp(N\sigma_{10}d)\,.$$

σ_{3914} is the total cross section for the production of 3914 band emission whether by electron capture or ionization, and Q is the "effective" cross section for the production of Balmer-α photons at a distance of 17.8 cm into the collision chambe More precisely,

$$Q = F_{3s}(z)\sigma_{3s} + F_{3P}(z)\sigma_{3p} + F_{3d}(z)\sigma_{3d}\,,$$

where $F_{3m} = 1 - \exp[-A(3m)z/v]$,

z is the distance of the observation region into the collision chamber, v is again the proton velocity, and $A(3m)$ is the total transition probability per unit time from the $3m$ state. For $z = 17.8$ cm, both F_{3p} and F_{3d} are nearly 1 for all v used so that

$$Q = F_{3s}\sigma_{3s} + (\sigma_{3p} + \sigma_{3d})\,.$$

Both Q and σ_{3914} are measured in separate experiments with the results shown in Fig. 5. σ_{10} is an average of several measurements[12] and is shown in Fig. 5 also. Equations (3.1)–(3.3) yield

$$\sigma_c(3p,B) + \sigma_c(3d,B) = \frac{\bar{N}_R}{N_HN_N}\frac{(C')^2}{C}\frac{nL}{e}\frac{\exp(n\sigma_{10}d)}{P_e}Q\sigma_{3914}$$

as the final relation between the emission cross section and experimentally measured or known quantities. The value of G may be obtained from this expression by multiplication with P_e.

C. Results

The final result for $\sigma_c(3p,B) + \sigma_c(3d,B)$ is shown in Fig. 5 where its relation in size and shape to the total emission cross sections, Q and σ_{3914}, and to σ_{10} is displayed. The data points are also shown in Fig. 6 where the random error is indicated. This error varies from 100% at velocities where the cross section is small to 25% near the peak and is too large to justify a display of any structural features in the cross section other than the single peak at 1.2×10^8 cm/sec.

The total estimated systematic error in the result is 35% at the lower velocities and 45% at the higher velocities. The estimated systematic error includes errors associated with the P-value approximations and neglect of $3s$ contribution as discussed previously; ionization gauge calibration (< 8%); errors in Q, σ_{3914}, and σ_{10} (≃15% each); and instrumental effects (≃10% total). Isotropic radiation is assumed. This assumption is based on the fact

TABLE I. Typical values of experimental parameters for one data set at $v = 1.5 \times 10^8$ cm/sec. The symbols are T effective integrating time for signal data; C beam charge collected during time T; N_A accidental coincidence counts during time T; N_R real coincidence counts during time T; I_B beam current; p target gas pressure; T' integrating time for single-channel data; C' beam charge collected during time T'; N_H single-channel counts from Balmer-α detector during time T'; and N_N single channel counts from 3914 detector during time T'.

$T \simeq 38$ min	$p = 1.1 \times 10^{-4}$ Torr	$C' = 4.29 \times 10^{-7}$ C
$C = 1.5 \times 10^{-5}$ C	$N_A + NR \simeq N_A \simeq 1.7 \times 10^5$	$N_H = 4.58 \times 10^5$
$I_B = 0.66 \times 10^{-8}$ A	$T' = 1$ min	$N_N = 3.34 \times 10^6$

N_R	
1330	
1270	$\bar{N}_R = 685$
580	
1368	Experimental standard deviation = 551
550	Theoretical standard deviation $\simeq (2N_A)^{1/2} = 584$
696	Standard deviation of the mean $\simeq 185$
−192	70% confidence limit $\simeq 213$
−140	
369	
1018	

that the total emissions of Balmer-α and N_2^+ first negative radiation are nearly isotropic[7] and the assumption that the radiation of the two final systems are uncorrelated events (see Sec. IV).

IV. CORRELATION BETWEEN FINAL-STATE EXCITATIONS

The significance of our result for $\sigma_c(3p, B) + \sigma_c(3d, B)$ is revealed by an examination of the correlation between final-state excitations of systems produced in electron-capture collisions.

Consider an electron-capture collision between a proton and a N_2 molecule in which the hydrogen atom is excited to the ith state and the N_2^+ molecule is excited to the jth state. The cross section for this process is $\sigma(i, j)$.

The total cross sections for the excitation of the ith state of hydrogen and the jth state of N_2^+ are, respectively,

$$\sigma(i) = \sum_j \sigma(i, j), \quad \sigma(j) = \sum_i \sigma(i, j),$$

and the total electron-capture cross section is

$$\sigma_{10} = \sum_{ij} \sigma(i, j).$$

In a large number of electron-capture collisions, the fractions of hydrogen atoms excited to the ith state and N_2^+ molecules excited to the jth state are, respectively,

$$P_i = \sigma(i)/\sigma_{10}, \quad P_j = \sigma(j)/\sigma_{10}.$$

In the following, we shall use the term "probability" to refer to the probability for an event conditional upon the occurrence of an electron-capture collision. Then, P_i and P_j may be regarded as the probabilities for exciting the indicated states of the final systems. The probability that the excitation of both the ith state of hydrogen and the jth state of N_2^+ results from a single collision is

$$P_{ij} = P_i P_j = \sigma(i)\sigma(j)/\sigma_{10}^2 \tag{4.1}$$

provided that the two excitations are independent. If the two excitations are not independent, one can write

$$P_{ij} = \gamma(i, j)\sigma(i)\sigma(j)/\sigma_{10}^2 \tag{4.2}$$

where

$$\gamma(i, j) \equiv P(j|i)/P_j \equiv P'(i|j)/P_i$$

and where $P(j|i)$ is the conditional probability that the jth state of N_2^+ is excited given that the ith state of hydrogen is excited and $P'(i|j)$ is the similar probability for exciting the ith state of hydrogen given that the jth state of N_2^+ is excited. Note that $\gamma(i, j)$ is necessarily non-negative. For $\gamma(i, j) = 1$, Eq. (4.2) reduces to Eq. (4.1) which has been obtained assuming that the two excitations are independent.

The cross section for simultaneous excitation of the ith state of hydrogen and the jth state of N_2^+ is

$$\sigma(i, j) = P_{ij}\sigma_{10} = \gamma(i, j)\sigma(i)\sigma(j)/\sigma_{10}. \tag{4.3}$$

When $\gamma(i, j) = 1$, the right-hand side of Eq. (4.3) is

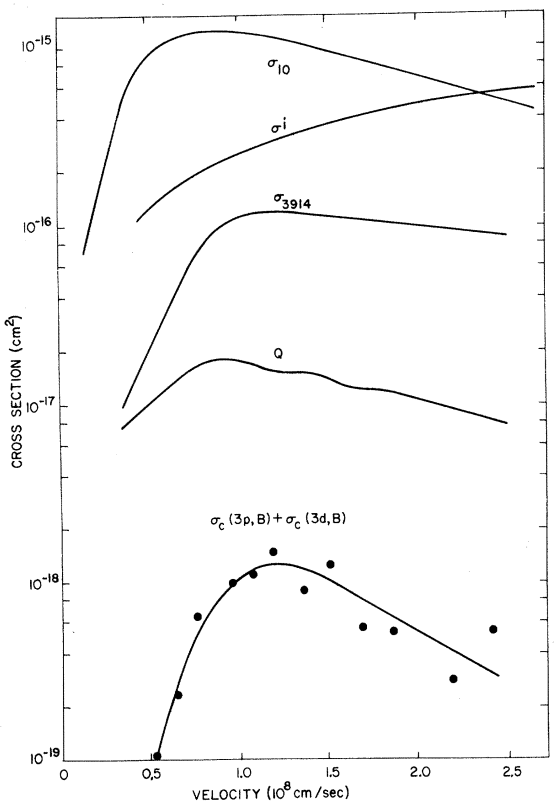

FIG. 5. Proton-nitrogen collision cross sections: σ_{10} total electron-capture cross section; σ^i total ionization cross section; σ_{3914} total cross section for the production of N_2^+ first negative (0, 0) photons; Q total cross section for production of Balmer-α photons at a distance of 17.8 cm into the collision chamber; $\sigma_C(3p, B) + \sigma_C(3d, B)$ cross section for production of Balmer-α (excluding the 3s state) and 3914 Å emissions from systems excited simultaneously in the same electron-capture collision.

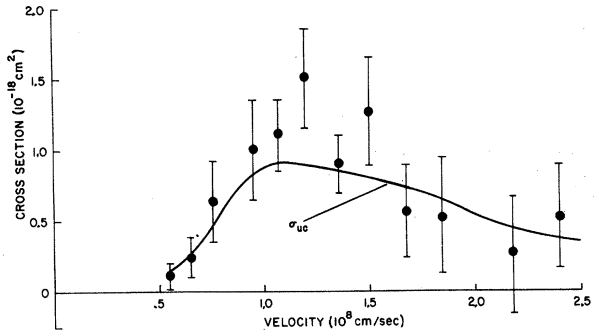

FIG. 6. Comparison of experimental and theoretical cross sections: σ_{uc} cross section calculated by assuming a lack of correlation between final-state excitations; $\sigma_C(3p, B) + \sigma_C(3d, B)$ experimental measurement. Error bars on experimental result indicate the 70% confidence limit for random errors.

the cross section for exciting the ith and jth states of the two systems when the excitations are independent. This quantity is denoted by $\sigma_{uc}(i, j)$ and is hereafter referred to as the "uncorrelated" cross section.

Values of $\gamma(i, j)$ other than unity account for varying degrees of correlation between the two excitations. For example, if the excitation of the ith state of hydrogen implies that the jth state of N_2^+ *must* be excited, then $\sigma(i) \equiv \sigma(i, j)$ and

$$\gamma(i, j) = \sigma_{10}/\sigma(j) , \tag{4.4}$$

and if the excitation of the jth state of N_2^+ implies that the ith state of hydrogen *must* be excited, then $\sigma(j) \equiv \sigma(i, j)$ and

$$\gamma(i, j) = \sigma_{10}/\sigma(i) . \tag{4.5}$$

If the excitation is such that $i \rightleftharpoons j$, then $\sigma(i, j) \equiv \sigma(i) \equiv \sigma(j)$ and

$$\gamma(i, j) = \sigma_{10}/\sigma(i, j) .$$

If the simultaneous excitation of the ith and jth states of the two systems is for some reason forbidden then $\gamma(i, j)$ is zero.

Although these special cases of correlation may be somewhat artificial, they do give an indication as to the possible values of $\gamma(i, j)$ for a particular pair of states (i, j) when $\sigma(i)$, $\sigma(j)$, and σ_{10} are known.

The function $\gamma(i, j)$ may be related to a correlation function, C, which takes on values from -1 to $+1$ by the correspondence: $\gamma = 0 \rightarrow C = -1$; $\gamma = 1 \rightarrow C = 0$; and $\gamma = \sigma_{10}/\sigma(i, j) \rightarrow C = +1$. A value of $\gamma(i, j)$ less than unity corresponds to a negative correlation between the indicated states while a value of $\gamma(i, j)$ greater than unity corresponds to a positive correlation.

So far the discussion has been concerned with the correlation between the excitation of the particular state i of hydrogen and the particular state j of N_2^+. A relation for the manifold of $\gamma(i, j)$ values resulting from a consideration of the dependences among the excitations of all the states of hydrogen with all the states of N_2^+ can be obtained. Summing both sides of Eq. (4.3) over all states of both systems, one obtains immediately

$$\sigma_{10} = \sum_{ij} \gamma(i, j)\sigma(i)\sigma(j)/\sigma_{10} . \tag{4.6}$$

But also, from the definitions of $\sigma(i)$, $\sigma(j)$, and σ_{10},

$$\sigma_{10} = \sum_{ij} \sigma(i)\sigma(j)/\sigma_{10} , \tag{4.7}$$

and hence upon subtraction of Eq. (4.6) from (4.7)

$$\sum_{ij} [1-\gamma(i,j)]\sigma(i)\sigma(j) \equiv 0 .$$

A value of unity for $\gamma(i,j)$ associated with every pair (i,j) is one obvious solution of the above identity. If $\gamma(i,j)$ is greater than unity for some pairs (positive correlation), then to satisfy the identity, $\gamma(i,j)$ must be less than unity (negative correlation) for other pairs.

The theoretically important situation in which $\gamma(i,j)$ is unity for all pairs (i,j) occurs if there is no correlation between any of the final-state excitations of the two systems. This means that in a large number of electron-capture collisions, the distribution over the possible final states of one system is independent of the specification of the final state of the other system and this distribution is a function of proton energy only according as the cross sections $\sigma(i)$ and $\sigma(j)$ vary with energy. A similar independence of final-state excitations is the condition found by Kessel and Everhart[13] for final *charge* states in Ar^+-on-Ar collisions except under certain conditions in which the inelastic energy loss, "Q," of the reaction displays a "triple-peaked" structure. In their work, the correlation between several final charge states of each of the product systems is determined and suggests strongly that there is a lack of correlation among all final charge-state excitations in the collisions they studied.

If it is assumed that $\gamma(i,j)$ is not equal to unity for all pairs (i,j) and that it has its strongest dependence on the energy defect, ΔE, associated with the (i,j) excitation, it might be reasonable to assume that $\gamma(i,j)$ decreases with increasing ΔE. Coupling this assumption with the fact that if some $\gamma(i,j)$ are greater than unity, then others must be less than unity, leads to the conclusion for endothermic reactions that those $\gamma(i,j)$ for which i and j are (or are near) the ground states must be greater than unity. However, an analysis of the Born-approximation data calculated by Mapleton[14] for simultaneous excitation of hydrogen and He^+ states in charge-exchange collisions of protons on He shows that the $\gamma(i,j)$ values for (i,j) near the ground state are unity to within 10% for most (i,j) pairs. Since the value of $\Delta E(i,j)$ varies most rapidly with i and j when i and j are near the ground state of the respective systems, one consequence of this analysis of Mapleton's data is that the $\gamma(i,j)$ do not depend on ΔE. It is shown in Appendix II that if the cross sections for the competing reactions depend exponentially on ΔE (such as is observed in the adiabatic region[15, 16], the excitations of the product systems are independent, i.e., $\gamma(i,j)$ is the same for all i and j (and thus equal to unity).

The modification of Eq. (4.3) to apply to the present investigation is

$$\sigma_c(3p,B)+\sigma_c(3d,B) = \gamma(\sigma_{3p}+\sigma_{3d})\sigma_{3914}{}^{10}/\sigma_{10} , \tag{4.8}$$

where $\sigma_{3914}{}^{10}$ is the total cross section for producing 3914 band emission in electron-capture collisions. Here, γ is an effective value for $3p$ and $3d$ excitations combined. The uncorrelated cross section [the right-hand side of Eq. (4.8) with $\gamma = 1$] may be written

$$\sigma_{uc} = \frac{(Q - F_{3s}\sigma_{3s})(\sigma_{3914}/\sigma_{10})}{1+\sigma_{3914}{}^{i}/\sigma_{3914}{}^{10}} ,$$

where $\sigma_{3914}{}^{i}$ is the total cross section for producing 3914 band emission in ionizing collisions. σ_{uc} is shown in Fig. 6 along with the experimental data points for $\sigma_c(3p,B)+\sigma_c(3d,B)$. The results of our measurements are used for Q and σ_{3914} (Fig. 5) and the result of Hughes *et al.*[17] for σ_{3s}. The estimate used here for the ratio $\sigma_{3914}{}^{i}/\sigma_{3914}{}^{10}$ is the ratio of total ionization and total electron-capture cross-section measurements reported by several investigators (see Fig. 5).[18] The significance of our experimental result is that the values of γ obtained from the data points for the measured cross section and this uncorrelated cross section by the relation

$$\gamma = [\sigma_c(3p,B)+\sigma_c(3d,B)]/\sigma_{uc}$$

are all unity to within experimental and approximation errors. The average of these 12 values of γ is 1.11 with a standard deviation of 0.34. That this value of γ indicates a lack of correlation of final-state excitations between the $n=3$ states of hydrogen and the B state of N_2^+ is evident from the fact that a positive correlation of the type given by Eq. (4.4) or (4.5) yields a value of γ more than an order of magnitude greater than unity and that for a correlation factor, C, of -1, $\gamma = 0$.

A measured value of $\gamma = 1$ for only one pair of final-state excitations in electron-capture collisions does not necessarily imply a lack of correlation among all excitations of the two systems. However, because the two states chosen for study here were selected rather arbitrarily, these results are strong evidence for a lack of correlation among all final states of the two systems. The results of Kessel and Everhart and Mapleton provide further confirmation of this hypothesis. However, even though these two works and our investigation consider three separate collision processes, in all three cases the energies of the impacting particles are well above the threshold energies for the excitation processes investigated. An examination of the degree of correlation closer to threshold may yield significantly different results.

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APPENDIX I

A serious problem encountered in our measurements is the extraction of a small real coincidence

signal from a large accidental coincidence noise. The real coincidence signal is $\dot{N}_R T$ where $\dot{N}_R$ is given by Eq. (2.3) and, T is the signal integration time. If ideal counting statistics apply, the noise is defined as

$$N = [(2\tau \dot{N}_H \dot{N}_N)\, T]^{1/2} ,$$

where $\dot{N}_H = (InL/e) A_1 Q$,

and $\dot{N}_N = (InL/e) A_2 \sigma_{3914}$.

The signal-to-noise ratio is then

$$\mathrm{STN} = \frac{\dot{N}_R T}{N} \simeq \sigma_C \left(\frac{A_1 A_2}{2Q\sigma_{3914}} \right)^{1/2} \left(\frac{P_e^{\,2}}{\tau} \right)^{1/2} T^{1/2} ,$$

where $\sigma_C = \sigma_C(3p, B) + \sigma_C(3d, B)$.

Reasonable estimates of the quantities appearing in this expression give that the integrating time required to obtain a given STN ratio in our experiment is

$$T = (\mathrm{STN})^2 \times 10^3 \text{ sec.}$$

The value of τ used in the experiment is chosen to maximize the function $(P_e^{\,2}/\tau)$. The length of observation region used is chosen to maximize the product ΩP_e, where Ω is the solid angle intercepted by the lenses and is a linear factor appearing in A_1 and A_2.

APPENDIX II

The energy defect of a collision may be written

$$\Delta E(i,j) = E_0 + E(i) + E(j) ,$$

where E_0 is a constant and $E(i)$ and $E(j)$ are the excitation energies of the two product systems. If the cross section for the competing excitations varies exponentially as

$$\sigma(i,j) = \sigma_0 e^{-\Delta E(i,j)/K} ,$$

then the terms representing the individual excitations of the two systems can be factored, giving

$$\sigma(i,j) = \sigma_0 e^{-E_0/K_e - E(i)/K_e - E(j)/K}$$

Thus, although the absolute value of $\sigma(i,j)$ varies with, say, i, the distribution over states j does not change with i, indicating a complete lack of correlation.

In general, if $\sigma(i,j)$ can be written as a product

$$\sigma(i,j) = F(i)G(j) ,$$

a lack of correlation results. The power-law expression[15,19]

$$\sigma(i,j) = \sigma_0 (K/|\Delta E(i,j)|)^n$$

cannot be put in this product form analytically, but may possibly be accurately approximated over a large number of final-state excitations by such a product function.

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Radiative Lifetimes of Excited Electronic States in Ionic-Species of Oxygen

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The beam-foil technique has been used to measure lifetimes of excited electronic states in oxygen II through VI. The possibility of cascading from above affecting the measurements is discussed. Only the most intense multiplets and members thereof were observed in the range between 2000 and 5000 Å. Where it is possible the lifetimes and some transition probabilities that may be extracted are compared with available calculations. A considerable number of transitions were untabulated and were identified through the use of a computer program.

I. INTRODUCTION

Research in plasmas and astrophysics is highly dependent on the oscillator strengths of radiative transitions from excited electronic states of neutral and ionic species of atoms. The absorption oscillator strengths, f_{ij}, are related to the radiative lifetimes, τ, of such states through the transition probability for spontaneous emission, A_{ij}:

$$A_{ij} = (8\pi^2 r_0 c g_i / g_j \lambda_{ij}^2) f_{ji} , \quad (1)$$

$$\tau_i = (\textstyle\sum_j A_{ij})^{-1} , \quad (2)$$

$$\tau_i = (8\pi^2 r_0 c g_i \textstyle\sum_j f_{ji} / g_j \lambda_{ij}^2)^{-1} , \quad (3)$$

where g is the statistical weight of the appropriate state and r_0, c, and λ are the classical electron radius, the velocity of light, and the wavelength of the transition, respectively. The beam-foil technique[1–4] has become a useful and quite productive method for the determination of radiative lifetimes and, when used in conjunction with other techniques, yields values for transition probabilities.

The beam-foil technique used in this determination of radiative lifetimes of oxygen has been described in detail previously[4] (applied to measurements in nitrogen) but is summarized briefly here. A high-velocity beam of singly charged ions from an accelerator is directed through a thin, freely supported foil of carbon (~ 1000 Å thick). The ions emerge from the foil in a variety of ionic species in a state of electronic excitation because of charge-loss interactions. The process of spontaneous decay from these states as the ions move along gives rise to optical radiation with intensity a function of distance from the foil. Observation of this decay of intensity of a particular spectral line yields the radiative lifetime of the upper state of the transition involved. For a decay where the upper state is populated only directly by the foil and not from any cascading from other excited states, the intensity may be described by

$$\ln I_\lambda = (\ln I_\lambda)_{t_0} - x/v\tau_i , \quad (4)$$

where x is the distance from the foil and v is the velocity of the beam particles.

II. EXPERIMENTAL PROCEDURE

The oxygen ion beam was provided by the Van de Graaff accelerator facility at the Air Force Cambridge Research Laboratories. The vertical orientation of this machine necessitated bending the beam 90° by an analyzing magnet. This magnetic analysis also assured the chemical purity of the beam since many gases were present in the sample bottle feeding the ion source. Excitation