# Excitation of low-energy H atoms in H + Ne collisions

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Excitation of 0.05–2.5-keV H atoms in H+Ne collisions has been examined by measuring the absolute emission cross sections for Balmer- $\alpha$ , Balmer- $\beta$ , and Lyman- $\alpha$  radiations resulting from the collisions. Lyman- $\alpha$  emission was also observed when an electric field was applied in the interaction region to investigate excitation of H atoms to the (metastable) 2s state. The radiation polarization for each of the above emissions was measured. A new method for absolute calibration of the Lyman- $\alpha$  detector is described, and Lyman- $\alpha$  emission cross sections for H<sup>+</sup>+Ar and  $e^-$ +H<sub>2</sub> collisions are compared with previous results, the latter reaction giving a significantly smaller cross section than generally accepted. The peculiar behavior of the H+Ne interaction indicated by the measurements is discussed.

## I. INTRODUCTION

In earlier publications,<sup>1-3</sup> the absolute emission cross sections and radiation polarizations for Balmer- $\alpha$  (H<sub> $\alpha$ </sub>) and Balmer- $\beta$  (H<sub> $\beta$ </sub>) emissions resulting from low-energy H collisions with Ar, He, H<sub>2</sub>, N<sub>2</sub>, and O<sub>2</sub> targets were reported for laboratory H energies between about 0.05 and 2.5 keV. This paper begins with a report in Sec. II of similar measurements for H + Ne collisions. The absolute calibration of the photon detector used for this work has been described earlier,<sup>1</sup> and will not be reviewed here.

However, because this is the first of a series of papers which will report Lyman- $\alpha$  ( $L_{\alpha}$ ) emission cross sections for various reactions, the  $L_{\alpha}$  detector and its absolute calibration are described in detail in Secs. III and IV. The cross sections for  $L_{\alpha}$  emission from H<sup>+</sup>+Ar and e<sup>-</sup>+H<sub>2</sub> collisions measured with this detector calibration are compared with other data in Sec. IV.

The results of the  $L_{\alpha}$  emission cross-section measurements for H + Ne collisions are presented in Sec. V. By analysis of these data and those for H<sub> $\alpha$ </sub> and H<sub> $\beta$ </sub> emission presented in Sec. II, the fractions of the total  $L_{\alpha}$  from direct excitation and from cascade population of the 2*p* state were determined.

Section VI describes the observations made of the  $L_{\alpha}$  emission from H + Ne collisions when a 700 V/cm electric field was applied to the interaction region. An analysis of these data resulted in a determination of the cross section for excitation to the (metastable) 2s state of H in the collisions.

The results of these various measurements are summarized in Sec. VII, where they are compared with results from collisions of H with other target species. An approximate determination of the polarization of  $L_{\alpha}$  emission from the electric-field-induced decay of the 2s state of H is also presented there.

The techniques used in these experiments to generate the H-atom beam and to measure the target-cell pressure have been described in earlier papers,<sup>1,4,5</sup> and will not be discussed here. All photon signals were measured as a function of target-cell pressure, and the normalized photon signals (i.e., the actual signals divided by the pressure) were extrapolated to zero pressure to eliminate the influence of second-collision processes occurring in the target cell.

For all reactions investigated, the polarizations of the emitted radiations were measured. The polarizations are defined by  $P = (I_{\parallel} - I_{\perp})/(I_{\parallel} + I_{\perp})$ , where  $I_{\parallel}$  and  $I_{\perp}$  are the emission intensities at 90° to the H-beam axis with their electric vectors parallel and perpendicular to the H-beam axis, respectively. These data were used to obtain total cross sections Q from the cross sections Q' measured for photon emission at 90° to the beam axis: Q = Q'(1 - P/3). In this paper, cross sections labeled Q' will always refer to measured (or calculated) data associated with photon emission at 90° to the H-beam axis.

# II. $H_{\alpha}$ and $H_{\beta}$ EMISSION CROSS SECTIONS

The measured total  $H_{\alpha}$  and  $H_{\beta}$  emission cross sections,  $Q_{\rm em}(H_{\alpha})$  and  $Q_{\rm em}(H_{\beta})$ , are plotted in Fig. 1 versus laboratory H-atom energy (E). For E > 0.2 keV, the cross sections  $Q_{\rm em}(ns \rightarrow 2p)$  and  $Q_{\rm em}(np \rightarrow 2s + nd \rightarrow 2p)$  for n = 3 or 4 identify the divisions of  $Q_{\rm em}(H_{\alpha})$  or  $Q_{\rm em}(H_{\beta})$ according to the indicated H-atom transitions causing the emissions. These divisions were determined by measuring the increases in the  $H_{\alpha}$  and  $H_{\beta}$  signals with increasing distance into the target cell due to the delayed decay of the long-lived ns states of H.<sup>1</sup> (The short-dashed curves labeled  $np \rightarrow 2s$  and  $nd \rightarrow 2p$  will be discussed in Sec. V.) The polarizations of the total  $H_{\alpha}$  and  $H_{\beta}$  radiations, used to determine the Q shown from the measured Q', are presented in Fig. 2.

The uncertainties in  $Q_{\rm em}({\rm H}_{\alpha})$  and  $Q_{\rm em}({\rm H}_{\beta})$  are  $\pm 13.4\%$  and  $\pm 13.7\%$ , respectively, for  $E \ge 0.1$  keV, increasing to about  $\pm 20\%$  at the lowest *E*. For E > 0.25 keV,  $Q_{\rm em}(3s \rightarrow 2p)$  and  $Q_{\rm em}(4s \rightarrow 2p)$  are uncertain by  $\pm 13.5\%$  and  $\pm 14.3\%$ , respectively, while  $Q_{\rm em}(3p \rightarrow 2s + 3d \rightarrow 2p)$  and  $Q_{\rm em}(4p \rightarrow 2s + 4d \rightarrow 2p)$  are about twice as



FIG. 1.  $H_{\alpha}$  and  $H_{\beta}$  emission cross sections for H + Ne collisions.

uncertain. These uncertainties were determined by quadrature combination of all individual uncertainties known to affect the measured data or its analysis, the largest of which is the  $\pm 10.2\%$  assigned to the absolute calibration<sup>1</sup> of the photon detector used. Typical uncertainties in the radiation polarizations are flagged in Fig. 2.

The most striking feature of the data in Fig. 1 is clearly the large contributions to the total  $H_{\alpha}$  and  $H_{\beta}$  emissions made by the  $ns \rightarrow 2p$  transitions for E > 0.32 keV. For no other target species (including the published results<sup>1-3</sup> for Ar, He, H<sub>2</sub>, N<sub>2</sub>, and O<sub>2</sub>, or the unpublished results<sup>6</sup> for Kr and Xe) has *ns*-state excitation been so dominant. That most of the H<sub> $\alpha$ </sub> and H<sub> $\beta$ </sub> emissions result from  $ns \rightarrow 2p$ transitions for E > 0.32 keV is consistent with the small radiation polarizations shown in Fig. 2, for  $ns \rightarrow 2p$  transitions must give unpolarized radiations (because of the



FIG. 2. Polarization of  $H_{\alpha}$  and  $H_{\beta}$  radiation from H + Ne collisions.



FIG. 3. Photon detector for observing  $L_{\alpha}$  emission.

spherical symmetry of *ns*-state wave functions). For most other targets, the radiation polarizations are larger.

This predominance of *ns*-state excitation has a direct bearing on what might be expected from a measurement of  $L_{\alpha}$  emission resulting from H + Ne collisions. For  $n \ge 3$ , these *ns* states decay preferentially to the 2*p* state, resulting in cascade contributions to the total  $L_{\alpha}$  emission. Because of the long radiative lifetimes of excited *ns* states (all  $\tau_{ns} \ge 1.6 \times 10^{-7}$  sec), the  $L_{\alpha}$  signal from such cascade processes should depend on the distance into the target cell where the signal is observed, just as for the case of H<sub> $\alpha$ </sub> and H<sub> $\beta$ </sub> emission from  $ns \rightarrow 2p$  transitions. Indeed, it is this very fact which has been used here to obtain an absolute calibration of the detector used to study  $L_{\alpha}$  emission from H + Ne collisions, as will be described in Sec. IV.

## III. THE $L_{\alpha}$ DETECTOR

While the  $L_{\alpha}$  detector took on several configurations during the course of these measurements, a typical configuration is shown in Fig. 3. Photons produced by H + Ne collisions entered the detector through a small square (or rectangular) aperture and were rendered into a parallel beam by the LiF lens. They then traversed an interference filter (with a 15-nm FWHM bandpass and peak transmission at about 124 nm) and an O<sub>2</sub> gas-cell filter (described below), and impinged on the photocathode of a solar-blind photomultiplier through its MgF<sub>2</sub> window. The photocathode was 11.5 cm from the H-beam axis, and the electron beam was 4.3 cm from the 0.4 cm diam entrance aperture to the target cell.

For the  $L_{\alpha}$  radiation polarization measurements, a LiF window was mounted at the Brewster angle in the position where the photomultiplier is shown in Fig. 3. The reflected intensities  $I_{||}$  and  $I_{\perp}$  were measured by rotating the window by 90° about the optical axis.

The photomultiplier was operated in a pulse-counting mode, its amplified output being shared by two counters gated on/off to correspond to the beam on/off cycles.<sup>1</sup> Because all known sources of background counts were continuous in time, subtraction of the counts accumulated

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by the counters provided a measure of the  $L_{\alpha}$  photons from the collisions being investigated.

The O<sub>2</sub> gas-cell filter consisted of two LiF windows separated by 1.00 cm. Cold-trapped O<sub>2</sub> gas flowed through the filter at a rate of about 3 cm<sup>3</sup>/sec, the O<sub>2</sub> pressure in the cell being held at about 720 Torr for most measurements. The absorption of ultraviolet light by O<sub>2</sub> between 105 and 135 nm was first studied by Watanabe *et al.*,<sup>7</sup> who found that the absorption coefficient was large except at seven narrow wavelength "windows" centered at 110.8, 112.7, 114.3, 115.7, 116.7, 118.7, and 121.6 nm, this last window being very close to the  $L_{\alpha}$  wavelength (121.57 nm). Numerous workers have followed the pioneering work of Fite and Brackmann<sup>8</sup> in using such filters for wavelength discrimination during  $L_{\alpha}$  emission cross-section measurements.

The use of a photomultiplier with a MgF<sub>2</sub> window and an interference filter in conjunction with an  $O_2$  gas filter in the present experiment effectively blocked transmissions through the four shorter-wavelength windows identified above. Transmissions through the 116.7 and 118.7 nm windows were also reduced significantly relative to that through the 121.6 nm window. Nevertheless, emissions other than  $L_{\alpha}$  could pass through the three longerwavelength windows, and each reaction studied had to be analyzed for possible contamination of the desired  $L_{\alpha}$  signal. For H + Ne collisions, the O<sub>2</sub> filter was not even needed, for the Ne spectrum is free of lines from 96 to 256 nm, and the Ne<sup>+</sup> spectrum from 46 to 168 nm,<sup>9</sup> so the interference filter alone isolated  $L_{\alpha}$ .<sup>10</sup> Thus, the properties of the  $O_2$  filter could be studied using H + Necollisions as a source of  $L_{\alpha}$ .

A plot of the relative filter transmission as a function of  $O_2$  pressure is shown in Fig. 4. The line curve was calculated from

$$S(P) = S_0 e^{-K_p(P/760)L}, \qquad (1)$$

where S(P) is the  $L_{\alpha}$  signal at O<sub>2</sub>-cell pressure P,  $S_0$  is the signal at P=0, L is the cell length (1.00 cm), P is the O<sub>2</sub> pressure (in Torr), and  $K_p$  (in cm<sup>-1</sup>) is the O<sub>2</sub> absorption coefficient fitted to  $K_p=0.269+0.000478P$ . This  $K_p$ agrees well with other data, for example, by Watanabe et al.<sup>7</sup> Thus a pure  $L_{\alpha}$  signal should increase by a factor of 1.79 when O<sub>2</sub> at 720 Torr is removed from the filter in the experiment, with any measured difference in this fac-



FIG. 4. Relative  $L_{\alpha}$  signal versus O<sub>2</sub> filter pressure.

tor indicating that detected radiations other than  $L_{\alpha}$  may be a problem.

The H + Ne reaction also allowed an estimate to be made of the transmission of Doppler-shifted  $L_{\alpha}$  by the O<sub>2</sub> filter. (While the detector's optical axis was at 90° to the H-beam axis, its viewing field accepted photons radiated at 90°±7° to the H-beam axis.) Comparison of the Henergy dependence of the  $L_{\alpha}$  signals measured with and without O<sub>2</sub> in the filter showed that a small H-velocitydependent factor (1.018±0.010, at E=2.5 keV) was needed to correct  $L_{\alpha}$  signals measured with O<sub>2</sub> in the filter.

With the  $O_2$ -gas lines to the filter disconnected, the entire  $L_{\alpha}$  detector shown in Fig. 3 could be moved parallel to the H-beam axis to observe  $L_{\alpha}$  as a function of distance x into the target cell for  $4 \le x \le 12$  cm. As indicated in Sec. II, the  $L_{\alpha}$  signal from H + Ne collisions was found to increase with increasing x, the largest and most accurately measurable signal increases occurring for  $1 \le E \le 2$  keV. How this signal increase with x was used to provide an absolute calibration of the  $L_{\alpha}$  detector is described next.

# IV. CALIBRATION OF THE $L_a$ DETECTOR

The  $L_{\alpha}$  signal from the cascade population of the 2p state of H in H + Ne collisions should be a function of distance x into the target cell. This can be written formally as<sup>11</sup>

$$\epsilon_{cas}(x) = \epsilon \sum_{nl} Q'_{em}(nl \to 2p)(1 - e^{-x/v\tau_{nl}}) , \qquad (2)$$
$$= \epsilon Q'_{cas}(2p) \left[ \frac{\sum_{nl} Q'_{em}(nl \to 2p)(1 - e^{-x/v\tau_{nl}})}{\sum_{nl} Q'_{em}(nl \to 2p)} \right] = S_{cas}F(x) , \qquad (3)$$

where  $S_{cas}(x)$  is the normalized photon signal, the actual signal (in sec<sup>-1</sup>) divided by the H flux (in sec<sup>-1</sup>), the Ne density (in cm<sup>-3</sup>), and the length of beam path (in cm) viewed by the detector, v is the H velocity (in cm/sec),  $\tau_{nl}$ 

is the radiative lifetime (in sec) of an *nl* state of H which can populate the 2*p* state via  $nl \rightarrow 2p$  cascade transitions,  $Q'_{\rm em}(nl \rightarrow 2p)$  is the Balmer-line emission cross section (in cm<sup>2</sup>) for the transition,  $Q'_{\rm cas}(2p)$  is the sum of such cross sections, i.e.,

$$Q'_{\rm cas}(2p) = \sum_{nl} Q'_{\rm em}(nl \rightarrow 2p) ,$$

 $\epsilon$  is the total efficiency of the  $L_{\alpha}$  detector,  $S_{cas}$  is the total normalized  $L_{\alpha}$  signal from cascade processes (the value at infinite x), i.e.,  $S_{cas} = \epsilon Q'_{cas}(2p)$ , and F(x) is the quantity in the brackets in Eq. (3). Note that F(x) is simply the fraction of the total normalized  $L_{\alpha}$  signal from cascade from all *nl* states observed at distance x into the target cell.

In contrast, because of the very short radiative lifetime of the 2p state  $(1.6 \times 10^{-9} \text{ sec})$ , the normalized  $L_{\alpha}$  signal from direct excitation to the 2p state should be effectively independent of x, i.e.,  $S_{2p} = \epsilon Q'_{ex}(2p)$ . Thus, if the total normalized  $L_{\alpha}$  signal  $S_m(x)$  vs x is measured and F(x)calculated,  $S_{2p}$  and  $S_{cas}$  can be determined by fitting the  $S_m(x)$  data to

$$S_m(x) = S_{2p} + S_{cas}F(x)$$
 (4)

From this result, the fraction  $f_{cas}$  of the total  $L_{\alpha}$  emission from cascade processes can be determined from

$$f_{\rm cas} = \frac{S_{\rm cas}}{S_{2p} + S_{\rm cas}} = \frac{Q'_{\rm cas}(2p)}{Q'_{\rm ex}(2p) + Q'_{\rm cas}(2p)} , \qquad (5)$$

the second equality following from the fact that each normalized  $L_{\alpha}$  signal must be in direct proportion to the corresponding emission cross section (i.e.,  $S = \epsilon Q'$ ). Thus, a knowledge of the various  $Q'_{\rm em}(nl \rightarrow 2p)$  used to find  $Q'_{\rm cas}(2p)$  and F(x), and a measurement of  $S_m(x)$ , allows the cross section  $Q'_{\rm ex}(2p)$  and the total  $L_{\alpha}$  emission cross section

$$Q'_{\rm em}(L_{\alpha}) = Q'_{\rm em}(2p) + Q'_{\rm cas}(2p)$$

to be determined.

The data presented in Fig. 1 clearly show that for  $1 \le E \le 2$  keV, the  $ns \rightarrow 2p$  transitions dominate the cascade population of the 2p state from the n = 3 and 4 levels. In order to estimate how much 2p-state population from higher-lying *ns* states might occur, the ratio of the cross sections for excitation to the 4s and 3s states was examined as a function of E. The result of this study is shown in Fig. 5, where the cross-section ratio  $Q_{ex}(4s)/Q_{ex}(3s)$  has been multiplied by  $(\frac{4}{3})^3$  for the comparison.<sup>12</sup> The parameter R plotted thus represents the departure of the cross-section ratio for *ns*-state excitation from the  $n^{-3}$  scaling law, which predicts that

$$Q_{\rm ex}(ns) \propto \frac{1}{n^3}$$
, (6)

as might be expected at high E. (Thus, if this law were to be followed here, the value of R would be unity.) As can be seen, R varies linearly with log E and extrapolates to zero at about the energy threshold for excited H production.

This rather surprising result led to examinations of R for the other H + target collisions investigated. While some of the data exhibited considerable scatter (because  $H_{\alpha}$  and  $H_{\beta}$  emission from *ns* states is generally not so dominant as for Ne targets), for all reactions, the R



FIG. 5. Scaled excitation-cross-section ratio for 3s and 4s states populated in H + Ne collisions.

shown in Fig. 5 fitted the data to within measurement uncertainties, even though the excitation cross sections themselves exhibited differences in both magnitude and dependence on E.

It was assumed that the same R values would apply to excitation-cross-section ratios for higher-lying *ns* states. Thus, for example, it was predicted that  $Q_{ex}(5s) = R(\frac{4}{5})^3 Q_{ex}(4s)$  could be determined to within an estimated  $\pm 50\%$  uncertainty for  $1 \le E \le 2$  keV, and that  $Q_{ex}(ns)$  could be determined for  $n \ge 6$  to within an estimated  $\pm 100\%$  uncertainty by successive step increases in *n*. While this assumption could not be tested for these higher-lying *ns* states, the cross section

$$Q_{\rm ex}(2s) = (1/R)(\frac{3}{2})^3 Q_{\rm ex}(3s)$$

predicted from this model was in good agreement with the measured result (as will be shown in Sec. VII), lending confidence to use of the assumption for  $n \ge 5$ .

Figure 1 shows that for  $1 \le E \le 2$  keV, cascade population of the 2p state from  $ns \rightarrow 2p$  transitions will be much larger than from  $nd \rightarrow 2p$  transitions, which are only a part of the measured  $Q_{em}(np \rightarrow 2s + nd \rightarrow 2p)$ . It was, however, desirable to have some estimate of the magnitude of cascade population of the 2p state from nd states. A study was thus made of the ratio of the excitation cross sections to the 4d and 3d states as a function of E for targets other than Ne  $(nd \rightarrow 2p$  transitions being the source of larger fractions of the H<sub>a</sub> and H<sub>b</sub> emissions for the other reactions). The fit of the data to the R plotted in Fig. 5 was poor when an  $n^{-3}$  scaling law was used, generally giving R values only about 60% as large as those plotted.

On the hypothesis that the excitation cross sections should be strongly dependent on the small-radius part of the excited-state H wave function, the scaling law

$$Q_{\rm ex}(nl) \propto \frac{(n+l)!}{(n-l-1)!n^{2l+4}}$$
(7)

was investigated. The quantity on the right of Eq. (7) is simply the square of the coefficient of the leading term of the *nl*-state radial wave function. For *ns* states, it gives the familiar  $n^{-3}$  scaling law of Eq. (6). For *np* states,

$$Q_{\rm ex}(np) \propto \frac{(n+1)(n-1)}{n^5}$$
, (8)

and for nd states,

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$$Q_{\rm ex}(nd) \propto \frac{(n+2)(n+1)(n-1)(n-2)}{n^7}$$
 (9)

For excitation to the 3d and 4d states, application of Eq. (9) gives  $R = 1.665Q_{ex}(4d)/Q_{ex}(3d)$ , if R is again defined as the departure of the cross-section ratio from this scaling-law prediction. With R determined in this way, the data for H impact on the various targets investigated generally did follow the dependence of R on E shown in Fig. 5 for E > 0.25 keV. While the quality of the fit was not as good as for *ns*-state excitation (for example, giving R values 11% above and 30% below that plotted for H + He and H + Ar collisions, respectively, at E = 1.6 keV), it was clearly better on average than that predicted by the  $n^{-3}$  scaling law and was used here for the estimates needed to evaluate *nd*-state excitation for H + Ne collisions in the same way that *ns*-state excitation was handled above.

The procedure used to analyze the  $L_{\alpha}$  emission from H + Ne collisions for the detector calibration is summarized below. It was first assumed that the measured  $Q'_{\rm em}(np \rightarrow 2s + nd \rightarrow 2p)$  for n = 3 and 4, similar to the  $Q_{\rm em}(np \rightarrow 2s + nd \rightarrow 2p)$  shown in Fig. 1 but not corrected for polarization, resulted entirely from  $nd \rightarrow 2p$  transitions. These  $Q'_{em}(nd \rightarrow 2p)$  were then used to estimate  $Q'_{\rm ex}(nd)$  for n=3 and 4, which were used together with the measured  $Q'_{ex}(ns)$ , the scaling laws from Eqs. (6) and (9), and the R from Fig. 5, to give a set of  $Q'_{em}(nl \rightarrow 2p)$ for all  $n \ge 3$ .  $Q'_{cas}(2p)$  and F(x) were calculated, and measured values of  $S_m(x)$  were used in Eqs. (4) and (5) to find  $Q'_{ex}(2p)$ . Assuming the scaling law in Eq. (8) for np-state excitation was valid (it could not be tested here), and again taking R from Fig. 5,  $Q'_{ex}(np)$  were estimated from this  $Q'_{ex}(2p)$ . This allowed  $Q'_{em}(np \rightarrow 2s)$  for n=3and 4 to be determined, and these results were subtracted from the measured  $Q'_{\rm em}(np \rightarrow 2s + nd \rightarrow 2p)$  to obtain new values for  $Q'_{\rm em}(nd \rightarrow 2p)$ . The entire process was then iterated until a reasonable convergence was obtained. The uncertainties assigned to the  $Q'_{em}(nd \rightarrow 2p)$  resulting from the analysis were estimated to be  $\pm 100\%$  of their determined values.

Three sets of measured  $S_m(x)$  data at E = 1.6 keV were used for the calibration. One set is shown in Fig. 6, along with the least-squares fit to the measured results. The values of  $S_{2p}$  and  $S_{cas}$  obtained from the fit and their un-



FIG. 6. Relative  $L_{\alpha}$  signal  $S_m(x)$  versus distance into target cell.

certainties are indicated. A value of  $f_{\rm cas} = 0.388 \pm 0.6\%$ resulted from these data, the other sets of  $S_m(x)$  data giving  $f_{\rm cas}$  values of  $0.389 \pm 0.8\%$  and  $0.389 \pm 0.7\%$ . The agreement among these values is very good, even though the data were taken at different times over the course of these studies, and with different  $L_{\alpha}$ -detector configurations.<sup>10</sup>

A summary of  $Q'_{em}(nl \rightarrow 2p)$  and their estimated uncertainties for E = 1.6 keV is presented in Table I, together with the fractions of the total cascade population of the 2p state resulting from the various transitions. As can be seen, the  $3s \rightarrow 2p$  and  $4s \rightarrow 2p$  transitions amount to about 90% of the total. The uncertainty assigned to the total  $Q'_{cas}(2p)$  represents a direct sum of the individual uncertainties in the various  $Q'_{em}(nl \rightarrow 2p)$  judged to be correlated, and a quadrature combination of those judged to be uncorrelated.

Using these results, the  $L_{\alpha}$  emission cross sections for H + Ne collisions at 90° to the H-beam axis at E = 1.6 keV were found to be the following:  $Q'_{cas}(2p) = 1.70 \times 10^{-17}$  cm<sup>2</sup> ±13.7%,  $Q'_{ex}(2p) = 2.68 \times 10^{-17}$  cm<sup>2</sup> ±14.0%,  $Q'_{em}(L_{\alpha}) = 4.38 \times 10^{-17}$  cm<sup>2</sup> ±13.9%, and  $Q'_{std}(L_{\alpha}) = 3.32 \times 10^{-17}$  cm<sup>2</sup> ±13.9%. This last cross section (the "measured" value at x = 4.3 cm into the target cell) served as a detector-calibration standard on a daily basis to place other measurements on an absolute scale, i.e., to find the detector-efficiency calibration constant

Transition $nl \rightarrow 2p$	$\frac{Q'_{\rm em}(nl \rightarrow 2p)}{(10^{-17} \rm \ cm^2)}$	$Q'_{\rm em}(nl \rightarrow 2p)$ Uncertainty	Fraction of total cascade		
$3s \rightarrow 2p$	1.292°	±13.5%°	0.759		
$4s \rightarrow 2p$	0.233°	±14.3 %°	0.137		
$5s \rightarrow 2p$	0.067	$\pm 50\%$	0.040		
$ns \rightarrow 2p^{a}$	0.045	$\pm 100\%$	0.026		
$3d \rightarrow 2p$	0.041	$\pm 100\%$	0.024		
$4d \rightarrow 2p$	0.013	$\pm 100\%$	0.008		
$nd \rightarrow 2p^{b}$	0.010	$\pm 100\%$	0.006		
$Q'_{\rm cas}(2p)$	1.701	$\pm 13.7\%^{d}$	1.000		

TABLE I. Cascade cross sections.

<sup>a</sup>Sum for  $n \ge 6$ .

<sup>b</sup>Sum for  $n \ge 5$ .

<sup>c</sup>Measured values from Fig. 1.

<sup>d</sup>See the text.



FIG. 7.  $L_{\alpha}$  emission cross sections for H<sup>+</sup>+Ar collisions. The data are from: •, present results; ×, Pretzer *et al.* (Ref. 13).

 $\epsilon = S_m$  (at x = 4.3 cm)/ $Q'_{\text{std}}(L_{\alpha})$  as needed.

In order to investigate whether the  $L_{\alpha}$ -detector calibration exhibited any dependence on E,  $S_m(x)$  data were also measured for E = 1.0, 1.25, and 2.0 keV, and analyses like that described above were made. If the relative value of  $\epsilon$ is set to 1.000 at E = 1.6 keV, the relative values at these other H energies were measured to be 1.003, 1.000, and 0.996, respectively. Considering that all relative  $\epsilon$  values cited are uncertain by more than  $\pm 2.8\%$  from the measurements alone, to say nothing about the dependence of the calculated  $Q'_{cas}(2p)$  and  $f_{cas}$  on E, this agreement was very satisfactory.

To compare this  $L_{\alpha}$ -detector calibration with other work, the  $L_{\alpha}$  emission cross section for H<sup>+</sup>+Ar collisions was measured at 90° to the beam axis and at x=4.3 cm into the target cell. The results are compared with those of Pretzer *et al.*<sup>13</sup> in Fig. 7. The agreement is well within the indicated uncertainties, a particularly satisfying result because this H<sup>+</sup>+Ar $\rightarrow$ L<sub> $\alpha$ </sub> reaction has been used as a standard by numerous workers to calibrate absolutely their own  $L_{\alpha}$  detectors (for example, by Birely and McNeal,<sup>14</sup> whose results for  $L_{\alpha}$  emission from H + Ne collisions will be shown later).

Another reaction frequently used as a standard for cross section measurements is  $e^- + H_2 \rightarrow L_{\alpha}$ . Fite and Brackmann<sup>8</sup> recognized early in their studies of this reaction that their photon detector (using an O<sub>2</sub>-gas filter and an I<sub>2</sub>-vapor photon counter, both with LiF windows, and calibrated absolutely by comparing  $L_{\alpha}$  emission from  $e^-$  + H collisions with Born-approximation calculations), could detect molecular  $H_2$  emissions in addition to  $L_{\alpha}$ . They thus measured only the cross section for countable ultraviolet (cuv) emission, reporting  $Q'_{\rm em}({\rm cuv})$ = $1.39 \times 10^{-17}$  cm<sup>2</sup> for 100 eV electrons. This value was subsequently revised to  $Q_{\rm gm}({\rm cuv}) = 1.48 \times 10^{-17}$ subsequently revised to  $Q_{em}(cuv) = 1.48 \times 10^{-17}$  cm<sup>2</sup>±3.6% by Kauppila *et al.*<sup>15</sup> Carrière and de Heer<sup>16</sup> studied the transmission of an O<sub>2</sub> filter by H<sub>2</sub> emissions (belonging to the Werner and Lyman bands), and estimated that about 80% of this cuv emission from  $e^- + H_2$  collisions was  $L_{\alpha}$ . Values of  $Q_{\rm em}(L_{\alpha})$  near  $1.2 \times 10^{-17}$  cm<sup>2</sup> have thus been adopted by most workers for detector calibrations.

It was thus with some surprise that the  $Q_{\rm em}({\rm cuv})$  obtained here was found to be  $8.49 \times 10^{-18} {\rm cm}^2 \pm 14.6\%$  for 100 eV electrons.<sup>17</sup> Furthermore, it was estimated that between about 5% and 25% of the observed cuv signal was still coming from H<sub>2</sub> emissions.<sup>18</sup> Using the  $L_{\alpha}$ emission fraction of  $85\pm10\%$  gives  $Q_{\rm em}(L_{\alpha})$ 



FIG. 8.  $L_{\alpha}$  emission cross sections for H + Ne collisions. The data points are from: •, present results for  $Q_m(L_{\alpha})$ ; ×, Birely and McNeal (Ref. 14). See the text for definitions of other cross sections shown.

=7.22×10<sup>-18</sup> cm<sup>2</sup>±18.8%. While this value agrees within uncertainties with the value  $8.2 \times 10^{-18}$  cm<sup>2</sup>±15% recently determined by Shemansky *et al.*,<sup>19</sup> it is only about 60% of the generally adopted result cited above. If the present  $Q_{\rm em}(L_{\alpha})$  and the  $Q_{\rm em}$  (cuv) of Kauppila *et al.*<sup>15</sup> are accepted, it appears that only about 50% of the cuv emission observed in the older studies was  $L_{\alpha}$ , and that cross-section data obtained using  $Q_{\rm em}(L_{\alpha})$  as a calibration standard will require downward revision.

# V. $L_{\alpha}$ EMISSION CROSS SECTIONS

The  $L_{\alpha}$  emission cross section for H + Ne collisions  $Q_m(L_{\alpha})$ , measured at a distance of x = 4.3 cm into the target cell, is shown by the data points plotted in Fig. 8. The cross-section values have been placed on an absolute scale using the  $Q'_{std}(L_{\alpha})$  discussed in Sec. IV, and have been adjusted to account for the measured  $L_{\alpha}$  polarization at x = 4.3 cm shown in Fig. 9. The values plotted are uncertain by  $\pm 14.5\%$  for  $E \ge 0.1$  keV, the uncertainty increasing to about $\pm 20\%$  at 0.04 keV. The data of Birely and McNeal<sup>14</sup> are shown for comparison. The agreement is not highly satisfactory, but the two sets of data are within mutual uncertainties where they overlap.

The other cross sections shown in Fig. 8 were determined from

$$Q_{\rm ex}(2p) = Q_m(L_a) - F(4.3)Q_{\rm cas}(2p)$$
 (10)



FIG. 9. Polarization of  $L_{\alpha}$  radiation from H + Ne collisions.

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and

$$Q_{\rm em}(L_{\alpha}) = Q_{\rm ex}(2p) + Q_{\rm cas}(2p)$$
 (11)

In these expressions,  $Q_{cas}(2p)$  is the cross section for the cascade population of the 2p state from  $nl \rightarrow 2p$  transitions, determined from the  $H_{\alpha}$  and  $H_{\beta}$  emission cross sections shown in Fig. 1 and the procedures discussed in Sec. IV for scaling the cascade cross sections from  $n \ge 5$  levels. F(4.3) is the fraction of this cascade cross section which results in observable  $L_{\alpha}$  emission at x = 4.3 cm into the target cell.  $Q_{ex}(2p)$  is the cross section for direct excitation to the 2p state, and  $Q_{em}(L_{\alpha})$  is that for total  $L_{\alpha}$  emission. The uncertainties in  $Q_{em}(L_{\alpha})$ ,  $Q_{ex}(2p)$ , and  $Q_{cas}(2p)$  are close to those cited above for  $Q_m(L_{\alpha})$ .

The short-dashed curves in Fig. 8 show the contributions made to  $Q_{cas}(2p)$  by  $ns \rightarrow 2p$  and  $nd \rightarrow 2p$  transitions for all  $n \ge 3$ . The  $ns \rightarrow 2p$  contribution for  $E \ge 0.2$  keV, and the  $nd \rightarrow 2p$  contribution for  $E \leq 0.2$  keV, again have uncertainties similar to those cited above. However, for E > 0.2 keV, the uncertainty in the  $nd \rightarrow 2p$  contribution increases rapidly and is about  $\pm 100\%$  of the contribution value for  $E \ge 1$  keV. This is because of the approximate procedures used to find the separate  $np \rightarrow 2s$  and  $nd \rightarrow 2p$ contributions to the measured  $Q_{em}(np \rightarrow 2s + nd \rightarrow 2p)$ shown in Fig. 1, for n = 3 and 4 (see Sec. IV). The results of this separation are shown by the short-dashed curves in Fig. 1. At low E, where the predicted  $np \rightarrow 2s$  contribution is very small, its large uncertainty has little influence on the  $nd \rightarrow 2p$  contribution. However, where the contributions have similar values, both are highly uncertain.

Note that  $Q_m(L_\alpha)$  and  $Q_{\rm em}(L_\alpha)$  are close in magnitude for  $E \leq 0.2$  keV. This is as it should be, for the bulk of the  $L_\alpha$  here results from  $Q_{\rm ex}(2p)$ , and what cascade population of the 2p state is present is dominated by  $nd \rightarrow 2p$ transitions, the *nd* states having fairly short radiative lifetimes. In contrast,  $Q_m(L_\alpha)$  and  $Q_{\rm ex}(2p)$  are approaching similar values at the higher E, most of the cascade here being from long-lived *ns* states, the fraction of which show up as  $L_\alpha$  emission at x=4.3 cm being small (e.g.,  $\sim 0.3$  at 2.5 keV).

## VI. 2s-STATE EXCITATION CROSS SECTION

 $L_{\alpha}$  emission from H + Ne collisions was also observed when the interaction region contained an electric field



FIG. 10. Apparatus for producing an electric field in a target cell.

parallel to the H-beam axis. The electric field was produced by applying potentials  $V_f$  and  $V_b$  to the center two electrodes shown in Fig. 10 (separated by 3.14 cm along the beam axis and centered at the distance x = 4.3 cm into the target cell). With this arrangement, it was necessary to examine the  $L_{\alpha}$  signal versus the electric-field magnitude to determine if the signal was "saturated" so that  $L_{\alpha}$ from only those H atoms excited in the detector's viewing field was being observed.

Electric fields of the same magnitude could be generated by any of the combinations of potentials: (a),  $V_f = 0$ and  $V_b = \pm 2W$ ; (b),  $V_f = \pm W$  and  $V_b = \mp W$ ; or (c),  $V_f = \pm 2W$  and  $V_b = 0$ . In general, combination (a) required larger W to achieve  $L_{\alpha}$ -signal saturation, because with  $V_f = 0$  there was no electric field between the first two electrodes (separated by about 1 cm) shown in Fig. 10. An electric field there served to prequench 2s atoms excited earlier along the H-beam path. The W required to achieve  $L_{\alpha}$ -signal saturation for the various potential combinations was consistent with calculations made using the 2s-state lifetime as a function of electric-field magnitude.<sup>20</sup> Potential combination (b) was finally adopted because of the smaller absolute values of  $V_f$  and  $V_b$  needed for any given electric field magnitude. However, for W = 1100 V, giving the 700 V/cm electric field used for most measurements, the measured  $L_{\alpha}$  signal and polarization were independent of the potential combination used.

It should also be noted that the various potential combinations discussed above gave electric fields both parallel and antiparallel to the H-beam axis. Although all data were taken with both electric-field directions, no measurable difference in either the  $L_{\alpha}$ -signal magnitude or the radiation polarization was ever observed.

The measured  $L_{\alpha}$  emission cross section for H + Ne collisions in a 700 V/cm electric field,  $Q_m(L_{\alpha})$ , is shown by the data points in Fig. 11. The cross-section values have been adjusted to account for the polarization of the emitted radiation shown in Fig. 12. The average uncertainty of the plotted data is  $\pm 15.3\%$ .

The other  $L_{\alpha}$  emission cross sections shown in Fig. 11 are related by



FIG. 11.  $L_{\alpha}$  emission cross sections for H + Ne collisions in a 700 V/cm electric field. The data points are from: •, present results for  $Q_m(L_{\alpha})$ ; ×, Birely and McNeal (Ref. 14). See the text for definitions of other cross sections shown.



FIG. 12. Polarization of  $L_{\alpha}$  radiation from H + Ne collisions in a 700 V/cm electric field.

$$Q_{\rm ex}(n=2) = Q_m(L_{\alpha}) - Q_{\rm cas}(n=2)$$
, (12)

$$= Q_{\rm ex}(2s) + Q_{\rm ex}(2p) . \tag{13}$$

Here,  $Q_{ex}(n=2)$  is the cross section for excitation to the n=2 level of H, and  $Q_{cas}(n=2)$  is the cross section for the cascade population of the n=2 level from all  $n \ge 3$ levels.  $Q_{ex}(2s)$  is the cross section for excitation to the 2s state, and  $Q_{ex}(2p)$  is again that for excitation to the 2pstate taken from Fig. 8, and assumed to remain unchanged by application of the electric field. It was assumed also that the radiative lifetimes of all excited states were sufficiently short in the 700 V/cm electric field that the  $L_{\alpha}$  signals observed from all processes would be independent of distance x into the target  $cell;^{21}$  hence  $Q_m(L_\alpha) = Q_{\rm em}(L_\alpha).$ 

Unfortunately,  $Q_{cas}(n=2)$  could not be determined here to high accuracy. While the cross sections for excitation to all *nl* states were estimated during the  $L_{\alpha}$ detector-calibration analysis discussed in Sec. IV, how the various  $m_l$  substates shared the *nl*-state populations could not be uniquely determined. In addition, only for the n=3 level were calculations available describing the branching ratios for  $nlm_1$ -substate decay to the n = 2 level in a 700 V/cm electric field. These calculations, made by Rouze et al.,<sup>22</sup> are summarized in Table II.

The  $Q_{cas}(n=2)$  shown in Fig. 11 was calculated using two approximations. First, it was assumed that the  $m_l$ substates of any given nl state were equally populated. Second, it was assumed that the branching ratios for decay of all  $nlm_1$  substates were the same as those for the n = 3 level given in Table II.<sup>23</sup> Fortunately, the resulting  $Q_{cas}(n=2)$ , which must be considered uncertain by about  $\pm 40\%$ , is small compared to  $Q_m(L_{\alpha})$ , so that the  $Q_{\rm ex}(n=2)$  determined from Eq. (12) is uncertain by only

TABLE II. Branching ratios for n = 3 to n = 2 transitions in

about  $\pm 18\%$ . This leads to an uncertainty in  $Q_{ex}(2s)$  of about  $\pm 30\%$  for  $E \ge 0.16$  keV.

The  $Q_{ex}(2s)$  determined by the above procedure may be compared with the measured values of Birely and McNeal<sup>14</sup> in Fig. 11. As can be seen, the present results are larger by about the same amount as shown in Fig. 8 for comparison of the  $L_{\alpha}$  emission cross section.

#### VII. DISCUSSION OF THE RESULTS

One of the interesting features of the data reported here for H-atom excitation in low-energy H + Ne collisions is the reduced importance of interactions leading to np- and nd-state populations for H-atom energies in the 1 keV region. Figure 1 shows that for the n = 3 and 4 levels, the ns states are much more heavily populated than other nl states. For no other target, including the other rare-gas atoms, or the N<sub>2</sub>, O<sub>2</sub>, and H<sub>2</sub> molecules,  $^{1-3,6}$  has ns-state excitation been so dominant. It is not that  $Q_{ex}(ns)$  is so large for Ne targets, but rather that  $Q_{ex}(nl)$  for the other states are unusually small. Figure 11 shows that  $Q_{ex}(2p)$ is also less than  $Q_{ex}(2s)$  in this range of E, a result also unique to Ne targets. Indeed, even for He targets, the cross sections for np- and nd-state excitation are several times larger than those for Ne (even though the ns-state excitation cross sections for He are only about 25% as large as those for Ne).

While this diminished importance of *np*- and *nd*-state excitation in H + Ne collisions is not understood, it has provided an opportunity to examine *ns*-state excitation in detail (in addition to providing a new  $L_{\alpha}$ -detector calibration). It was noted earlier<sup>6</sup> that the ratio of the 4s- to 3sstate excitation cross sections was close to the prediction of an  $n^{-3}$  scaling law for all rare-gas-atom targets. This has now been quantified, as shown by the data in Fig. 5, where the parameter R has been defined by the departure of this cross-section ratio from the  $n^{-3}$  law prediction. As noted in Sec. IV, a similar plot (although with increased data scatter) of this cross-section ratio for all other targets investigated yields the same dependence of R on Ε.

It was also noted in Sec. IV that if the scaling laws for np- and nd-state excitation given by Eqs. (8) and (9) are used, and R is again defined as the departure of the  $Q_{\rm ex}(4l)/Q_{\rm ex}(3l)$  ratio from the scaling-law predictions, a generally similar dependence of R on E is consistent with the data for targets other than Ne. Analyses of the  $H_{\alpha}$ emissions from decay of the 3s, 3p, and 3d states like that

TABLE III. Ratio of measured to predicted  $Q_{ex}(2s)$ .

a 700 V/cm electric field [data are from Rouze <i>et al.</i> (Ref. 22)].		data are from Rouze et al. (Ref. —	E (keV)	Measured $Q_{ex}(2s)$	
	Upper level substrate	Branching ratio to $n=2$ level	2.50	0.93	
	350	0.416	1.60	1.00	
	$3p_0$	0.263	1.25	1.00	
	$3p_{\pm 1}$	0.341	1.00	1.07	
	$3d_0$	0.622	0.80	1.11	
	$3d_{\pm 1}$	0.372	0.63	1.14	
	$3d_{\pm 2}$	0.982	0.50	1.30	



FIG. 13. Polarization of  $L_{\alpha}$  radiation from H + Ne collisions as a function of electric field.

discussed here for Ne targets have now been completed. If it is assumed that the total  $H_{\alpha}$  emission from decay of these states has been properly partitioned, the parameter R and the scaling laws assumed can be used to predict the total  $H_{\beta}$  emission from the various collisions as a function of E. The ratios of these predicted values to the measured results, when averaged over the data from  $0.25 \le E \le 2.5$ keV, are the following: 0.90 for He; 1.07 for Ar, N<sub>2</sub>, and O<sub>2</sub>; 1.09 for Kr; and 1.31 for Xe.<sup>24</sup> Considering the large differences among these targets in the magnitudes and Henergy dependences of these cross sections, the quality of these predictions is remarkable.

In order to examine whether the parameter R was unique to the n=3 and 4 levels of H, the cross section for excitation to the 2s state was predicted from

$$Q_{\rm ex}(2s) = (1/R)(\frac{3}{2})^3 Q_{\rm ex}(3s)$$

and is compared with the data (from Fig. 11) in Table III for  $E \ge 0.5$  keV. While there is some trend in the measured to predicted cross-section ratio with E, it is again remarkable that this simple model could make predictions of this quality. (While the data for 2s-state excitation for the other rare-gas atom and molecular targets have not yet been finally analyzed, preliminary considerations suggest that generally similar results will be obtained.)

It is clear that the parameter R and the scaling laws used in the analysis described in this paper apply to many different H-atom-excitation processes. It would be difficult to conclude that their application is only accidentally valid.

The high positive polarization of the  $L_{\alpha}$  radiation re-

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sulting from H + Ne collisions for  $E \le 0.5$  keV (see Fig. 9) suggests that most of the 2*p*-state-excited H atoms are here being produced in the  $m_l=0$  substate.<sup>25</sup> (This is another example of the uniqueness of Ne as a target, for  $L_{\alpha}$  radiation from other H + target collisions is less polarized.) The rapid decrease of the polarization at higher E may be due in part to the importance of cascade population of the 2*p* state from higher *ns* states, which should give unpolarized  $L_{\alpha}$ .

The polarization of the  $L_{\alpha}$  radiation from H + Ne collisions in a 700 V/cm electric field (see Fig. 12) is near zero for  $E \ge 0.2$  keV (although it increases at low E where an increasing amount of the  $L_{\alpha}$  comes from 2p-state excitation). This fact, however, is an artifact of using an electric field of 700 V/cm magnitude. While not part of the main theme of this paper, the polarization of  $L_{\alpha}$  from H + Ne collisions was studied as a function of electric-field magnitude. The result of this study is shown in Fig. 13.

For E = 0.4 keV with the electric field on, about 67% of the total observed  $L_{\alpha}$  results from excitation to the 2s state. The polarization of  $L_{\alpha}$  from electric-field quenching (i.e., 2s- and 2p-state mixing) of this state was calculated by Crandall<sup>26</sup> as a function of electric-field magnitude, and is shown as the short-dashed curve labeled P(2s) in Fig. 13. The short-dashed curve labeled  $P(L_{\alpha})$ is the  $L_{\alpha}$  polarization measured here with no electric field (which was assumed to remain unchanged when the electric field was applied<sup>27</sup>). Thus for an electric field above about 200 V/cm, the curve labeled P=0.67P(2s) $+0.33P(L_{\alpha})$  is approximately what would be expected from the indicated relative weighting of the other curves, and may be compared with the measured total  $L_{\alpha}$  polarization as a function of electric field shown by the data points. As can be seen, the agreement is generally satisfactory, supporting the validity of both the measured and calculated results.

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- <sup>10</sup>In fact, some data for H + Ne collisions were taken without including even the interference filter, using the rapid decrease in the photocathode quantum efficiency above 150 nm as the only wavelength discrimination.

- <sup>11</sup>These expressions are used for purposes of exemplification only. More complicated expressions, including the effect of the pressure gradient near the target-cell entrance and the pressure in the vacuum tank prior to the target cell, were used for data analysis. These expressions also assume that the 2pstate lifetime is zero, a good approximation here.
- <sup>12</sup>These  $Q_{ex}(ns)$  were assumed to be given by the measured  $Q_{em}(ns \rightarrow 2p)$  divided by the  $ns \rightarrow 2p$  branching ratios, i.e., cascade to these *ns* states from still higher-lying *np* states was neglected. This is a good approximation because these highlying *np* states decay very preferentially to the 1s and 2s states. Note also that for all *ns* states, cross sections Q=Q', because decay of these states must give unpolarized radiation.
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examined this problem in some detail, and a variety of experimental tests of the data. For example, the measured cuv signal increased by about 450% when  $O_2$  at 720 Torr was evacuated from the filter. However, for  $O_2$ -filter pressures above 500 Torr, the cuv signal exhibited the same dependence on pressure shown in Fig. 4 for pure  $L_{\alpha}$ , suggesting that most of the cuv signal observed here was  $L_{\alpha}$ .

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- <sup>21</sup>This should be a reasonable approximation, because all nl states become mixed in such a high electric field and assume the short radiative lifetimes of np states.
- <sup>22</sup>N. Rouze, C. C. Havener, W. B. Westerveld, and J. S. Risley, private communication. The data tabulated ignore offdiagonal terms in the density matrix. These are expected to be small.
- <sup>23</sup>This was estimated to be a better approximation than ignoring cascade from all  $n \ge 4$ . If cascade from  $n \ge 4$  is ignored, the  $Q_{cas}(n=2)$  values decrease by about 25%.
- <sup>24</sup>The value for Xe may be in error because of possible spectral contamination of the  $H_{\alpha}$  by Xe radiations.  $H_2$  was not included because of the unknown amount of target emission observed. The value for Ne would be 1.00, because it was fitted to the model.
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