

## Polarization of rare-gas radiation in the vacuum-ultraviolet region excited by electron impact: Helium and neon

P. Hammond,\* W. Karras, A. G. McConkey, and J. W. McConkey  
*Department of Physics, University of Windsor, Windsor, Ontario, Canada N9B 3P4*

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Vacuum-ultraviolet (vuv) polarization data are presented for the integrated radiation emitted following electron-impact excitation of He and Ne in the energy range from threshold to 500 eV. The data represent an improvement both in accuracy and energy range over those available previously. They can be used as secondary standards for calibrating vuv polarization analyzers and other optical devices.

### I. INTRODUCTION

The development of secondary standards<sup>1-3</sup> for absolute calibration in the vuv has highlighted the need for accurate polarization data for electron-impact excited vuv lines and for accurate measurements of the polarization sensitivity of detection equipment. In addition, electron-photon coincidence experiments,<sup>4-10</sup> in which polarization correlation functions are measured to allow extraction of the fine details of the excitation processes, have stimulated interest in linear and circular polarization measurements in the vuv region. Although polarization measurements are straightforward in the visible spectral region where transmission optics may be used, this is not the case in the vuv region, where reflection-type analysis must be used and where deterioration of surfaces with consequent changes in optical performance is to be expected. Thus there is a need for sources which emit radiation of accurately known polarization and which are convenient and simple to use for calibration purposes.

Electron-impact sources are attractive in that they are small and readily assembled, and, in fact, these are now widely used for intensity calibration of optical equipment<sup>1-3</sup> in this and other laboratories. However, few accurate measurements of vuv polarizations from those sources are available. This is highlighted by the situation in helium which is the most widely studied electron-impact source. In 1977, Standage<sup>11</sup> reviewed the existing polarization data for He and highlighted the large discrepancies which existed between the data from different laboratories. He determined some vuv line polarizations (for the 58.4- and 53.7-nm lines) using electron-photon coincidence data together with differential cross-section data. Because of the roundabout nature of this procedure, the errors were large ( $\sim 15\%$ ). Five years later Steph and Golden<sup>12</sup> updated this work using new coincidence data and were able to reduce the errors by about a factor of 2. The only other He data available in the literature are those of Mumma *et al.*,<sup>13</sup> who measured the angular distribution of the in-

tegrated ( $n\ ^1P \rightarrow 1\ ^1S$ ) radiation with a limited accuracy of about 5%. Their measurements covered the energy range from 25 to 150 eV. A very limited amount of vuv polarization data from electron beam sources using other targets is available.<sup>14</sup>

The present paper presents accurate data for the polarization of integrated radiation from He and Ne using a reflection analyzer with gold surfaces. Planned future publications will present similar information for the heavier rare gases (Ar, Kr, and Xe), for the 121.5-nm multiplet of He<sup>+</sup> and for the molecular targets H<sub>2</sub> and N<sub>2</sub>. The energy range is chosen so that the energy where the polarization passes through zero can be accurately assessed. This is important for various reasons as discussed later but not least from the point of view that operation of an electron-impact source at this energy with this source gas automatically provides an unpolarized radiation source which can then be used for polarization calibration of spectroscopic diagnostic equipment. Preliminary reports of some aspects of this work have been presented elsewhere.<sup>15-17</sup>

### II. BASIC THEORY

The basic groundwork, relative to the polarization of atomic line radiation excited by electron impact, was laid in the classic paper of Percival and Seaton.<sup>18</sup> This paper exposed the limitations of earlier treatments and provided formulas for line polarizations as a function of electron-impact energy in terms of magnetic sublevel cross sections and appropriate coefficients. The threshold selection rule  $\Delta M = 0$ , which is based on conservation of angular momentum arguments, allows the threshold polarization to be obtained directly. The depolarizing effects of fine or hyperfine interactions were also dealt with by these authors. The situation is straightforward if the fine or hyperfine separations are either small or large compared to the natural linewidth.

Blum<sup>19</sup> gives the following expression for the threshold polarization  $P_T$ :

$$P_T = \frac{(\frac{15}{2})^{1/2}(-1)^{L_2}G(L)_2 \begin{Bmatrix} 1 & 1 & 2 \\ L & L & L_2 \end{Bmatrix} \begin{Bmatrix} L & L & 2 \\ 0 & 0 & 0 \end{Bmatrix}}{\frac{1}{\gamma} \frac{2}{3(2L+1)} + (\frac{5}{6})^{1/2}(-1)^{L_2}G(L)_2 \begin{Bmatrix} 1 & 1 & 2 \\ L & L & L_2 \end{Bmatrix} \begin{Bmatrix} L & L & 2 \\ 0 & 0 & 0 \end{Bmatrix}}, \quad (1)$$

where  $L$  and  $L_2$  represent the orbital momentum of the upper and lower states of the observed decay,  $\gamma$  is the lifetime of the state in question and  $[G(L)]_2$  is a perturbation coefficient which takes account of fine and/or hyperfine depolarizing effects. For the situation where both fine and hyperfine splittings are larger than the line width and where the observation time is long compared to the lifetime, so that the time-dependent part of  $[G(L)]_2$  drops out, we have

$$G(L)_2 = \frac{1}{\gamma} \frac{1}{(2S+1)(2I+1)} \sum_{J,F} (2J+1)^2(2F+1)^2 \begin{Bmatrix} J & F & I \\ F & J & 2 \end{Bmatrix}^2 \begin{Bmatrix} L & J & S \\ J & L & 2 \end{Bmatrix}^2, \quad (2)$$

where the quantum numbers  $S, L, J, I, F$  have the usual meaning and relationships. Polarizations are difficult to measure in the near-threshold region not only because of low radiation intensities and the influence of cascade but also because of the perturbing effect of resonances in this energy region.<sup>20</sup> Comparison with theoretically predicted threshold values is often limited to extrapolated experimental data.

Heddle<sup>21-23</sup> has extended earlier work by McFarlane<sup>24</sup> on the application of the Bethe approximation to the polarization of impact radiation and has shown the following. First the energy  $E_p$  at which the polarization goes to zero is given (in this approximation) for optically allowed and forbidden excitations, respectively, by

$$E_p = e^3 R / 4C_j \quad (3a)$$

$$= 3BR, \quad (3b)$$

where  $R$  is the Rydberg constant and  $C_j$  and  $B$  are parameters which occur in the Bethe approximation and which have been calculated in some instances. We note that  $C_j$  is related to the intercept  $E_0$  of the so-called Fano plot of  $QE$  versus  $\ln E$  with the energy axis

$$E_0 = R / 4C_j. \quad (4)$$

where  $Q$  is the excitation cross section.

Heddle has investigated the behavior of  $E_p/E_0$  for a variety of targets and finds it to be almost universally close to  $e^3$  as predicted. Heddle<sup>23</sup> also shows that, if the polarization is plotted against  $\ln E$ , then the gradients  $G'$  of the curves at the energy  $E_p$  are given, for the optically allowed and optically forbidden excitation processes, respectively, by

$$G'_A(1+\beta) = \frac{-P_T}{6-2P_T}, \quad (5a)$$

$$G'_F(1+\beta) = \frac{-3P_T}{6-2P_T}. \quad (5b)$$

$\beta$  is the ratio of cascade to direct excitation cross sections at energy  $E_p$ . Heddle found rather good agreement between values of  $P_T$  deduced from the measured polarizations of 24 visible transitions using Eqs. (5a) and (5b) and those calculated using the theory of Percival and

Seaton.<sup>18</sup> It is of interest in the context of the present work to check the level of agreement which is obtained with the much more energetic vuv transitions.

A number of calculations<sup>24-28</sup> of the variation of  $P$  with impact energy have been carried out in a variety of approximations for He as target gas. To the authors' knowledge no such information is available for the other targets.

### III. EXPERIMENT

The apparatus consisted of crossed electron and gas beams in one chamber and a reflection polarization analyzer in a differentially pumped second chamber. The electron gun was designed to provide a well-collimated constant current beam over a wide energy range. A beam current of  $25 \mu\text{A}$  with an energy spread of 500 meV was typically used. A layer of conetic shielding enclosed the electron gun and interaction region to minimize the effect of stray magnetic fields. Energy calibration of the system was obtained by observing the onsets of the various emissions and comparing these with the spectroscopic values. Contact potentials were typically a few tenths of an eV.

Radiation from the interaction region passed through an aperture in the wall between the two chambers, traversed the reflection analyzer, and was detected using a channel electron multiplier (Galileo 4039-C). For the targets whose resonance radiation lay in the wavelength region above 100 nm, the detector cone was coated with CsI to enhance its sensitivity. Two different polarization analyzers were used. For most of the data accumulation, a simple single-reflection polarizer with an angle of incidence of  $57.5^\circ$  was used. The optical element was a 1-in.-diam, optically flat, gold-coated Pyrex mirror (Janos Optical Corporation). It was found that some deterioration of the surface quality occurred over a period of time such that the efficiency  $\eta$  of the polarizer changed with time. In addition, because of inherent inaccuracies in the measured optical constants for gold (see Khakoo *et al.*<sup>29</sup>) there was a consequent significant uncertainty (a few percent) in the actual value of  $\eta$  even assuming a perfectly clean, flat surface. If the reflection coefficients of the surface are  $R_s$  and  $R_p$  for the electric vector parallel and perpendicular to the plane of incidence, respectively, then  $\eta$  is defined as

$$\eta = \frac{R_s - R_p}{R_s + R_p} = \frac{R_s/R_p - 1}{R_s/R_p + 1} \quad (6)$$

and the polarization of the radiation is given, in terms of the measured polarization  $P'$ , by

$$P = P' / \eta . \quad (7)$$

For a multiple mirror device with  $n$  mirrors and the same angle of incidence in each case, Eq. (6) must be modified to

$$\eta = \frac{(R_s/R_p)^n - 1}{(R_s/R_p)^n + 1} . \quad (8)$$

Clearly, the larger  $n$  is the less significant any changes in  $R_s/R_p$  become and the closer to unity the correction factor of Eq. (7) becomes. For example, consider typically that  $R_s/R_p = 5$ , then a 10% change or error in this value would produce less than a 0.2% error in  $P$  if  $n = 4$ . Thus in order to place the polarization data on an absolute scale we used a three-mirror polarizer<sup>30,31</sup> in series with the single-mirror device. The polarization curves were thus fixed at two electron energies, 30 and 80 eV, for each target gas. Data taken with the single-mirror device were scaled to these values. In actual practice the angles of incidence onto the four mirrors were not all identical so  $\eta$  had to be evaluated using the relevant values of  $R_s$  and  $R_p$  for each mirror. The final value for  $\eta$  was 0.997.

The orientation of the analyzer was controlled using a stepping motor under computer control. Thus complete polarization ellipses could be plotted or, more usually, data could be collected at the four orthogonal positions of the analyzer corresponding to detection of  $I^{\parallel}$  or  $I^{\perp}$ , where these parameters refer to light intensities polarized parallel and perpendicular, respectively, to the electron beam (quantization) axis. The polarization of the radiation is defined in the usual way by  $(I^{\parallel} - I^{\perp}) / (I^{\parallel} + I^{\perp})$ . Data from the detector were routed into the memories of a multichannel analyzer. Automatic scanning of electron beam energy could be carried out so that the variation of  $I^{\parallel}$  and  $I^{\perp}$ , and hence  $P$ , as a function of impact energy could be obtained directly. Any slight residual misalignment of the analyzer was taken account of by averaging the two  $I^{\parallel}$  and  $I^{\perp}$  count rates.

Studies of the variation of  $P$  with beam pressure were carried out to ensure freedom from depolarizing effects such as imprisonment of resonance radiation. In He this meant that the background pressure in the system increased by no more than  $10^{-7}$  torr when the gas beam was operational.

An important point in any measurement of polarization is adequately accounting for background effects. In our case we had two possible sources of background. First, due to the very low pressures used there was a background contribution to the measured signal from the ambient background gas in the system. This was essentially unpolarized and was mainly due to background  $N_2$  or  $H_2O$ . This contribution could be established by shutting off the gas beam and monitoring the radiation from the background gas. There was also some impurity introduced along with the target gas despite the presence of a

cold trap in the gas line and an all-metal gas regulator. This appeared as a small background contribution at energies below the excitation threshold of the target species. By comparing the below-threshold signals with and without the gas beam running we were able to establish the relative background contributions from the two different sources. The contributions were roughly comparable in magnitude. It was assumed that similar relative background contributions were present at other energies and so the background taken with no gas beam was scaled up accordingly. At 50 eV in He the total background amounted to 1.5% of the total signal.

#### IV. ERRORS

Statistical errors were much less than 1% (except close to threshold) and hence the main component of the errors listed in the results section comes from uncertainty in the background subtraction discussed above. This led to a possible systematic error in the polarization of  $\pm 0.005$  at 80 eV when considering He as a target. We believe that systematic errors due to mechanical misalignment, etc. are rendered negligible by the averaging procedure mentioned above. As a check on this we measured the polarization of the radiation from an  $N_2$  target ( $\lambda > 105$  nm) (which we believe should be zero for incident electron energies between 100 and 500 eV), and found this to be  $0.000 \pm 0.005$ .

One caution which should be considered when our data are being used by other authors is that slight variations in the data may occur (depending on the particular channel electron multiplier being used) due to differing wavelength sensitivity variations. Since, in He for example, light covering the spectral range from 50.5 to 58.4 nm is being detected and since both the reflection coefficients of gold and the detection sensitivity of the channel electron multiplier may vary significantly over this spectral range, one might expect to measure slightly different polarizations for the integrated radiation if the polarizations of the different spectral features are different. We found, for example, that at 80 eV the measured polarization of the integrated radiation from He would be in the range 0.395–0.425 depending on the particular detector being used and on whether it had been coated with CsI. This factor is not included in the systematic errors quoted.

#### V. RESULTS AND DISCUSSION

The polarization data for the two gases are shown in Figs. 1–3 and are tabulated in Table I. Table II presents relevant parameters derived from the data. We shall discuss the data for the individual targets separately.

##### A. Helium

The polarization, Fig. 1, is observed to fall from a high value at threshold to a minimum around 25 eV. The threshold value of 0.78 is consistent with a true threshold value of unity, as would be predicted from the threshold angular momentum selection rules, given that the energy resolution of the  $e$  beam is approximately 500 meV. The curve rises again, to a maximum value around 37 eV, and

TABLE I. Polarization data for He and Ne.

Energy (eV)	Helium		Neon	
	Pol.	Stat. Error	Pol.	Stat. Error
$E_T$	0.783	0.060	0.065	0.023
18			0.009	0.013
20			0.152	0.004
22	0.727	0.016	0.229	0.003
24	0.510	0.005	0.267	0.002
26	0.497	0.003	0.298	0.003
28	0.524	0.004	0.318	0.003
30	0.533	0.003	0.344	0.001
35	0.568	0.004	0.356	0.002
40	0.572	0.002	0.374	0.001
50	0.541	0.001	0.365	0.001
60	0.501	0.001	0.331	0.001
70	0.462	0.001	0.286	0.001
80	0.421	0.001	0.251	0.001
90	0.382	0.001	0.212	0.001
100	0.358	0.001	0.186	0.001
120	0.296	0.001	0.145	0.001
140	0.252	0.001	0.108	0.001
160	0.214	0.001	0.089	0.001
180	0.181	0.001	0.066	0.001
200	0.151	0.001	0.057	0.001
220	0.130	0.001	0.045	0.001
240	0.109	0.001	0.033	0.001
260	0.087	0.001	0.020	0.002
280	0.065	0.001	0.015	0.001
300	0.046	0.001	0.011	0.001
320	0.036	0.001	0.001	0.001
340	0.028	0.001	-0.002	0.001
360	0.009	0.001	-0.009	0.001
380	-0.003	0.001	-0.012	0.001
400	-0.012	0.001	-0.017	0.001
420	-0.026	0.001	-0.019	0.001
440	-0.033	0.001	-0.023	0.001
460	-0.043	0.001	-0.032	0.001
480	-0.049	0.001	-0.040	0.001

then falls off rapidly crossing the  $P=0$  axis at  $375 \text{ eV} \pm 10 \text{ eV}$ . Figure 1 also shows a variety of theoretical calculations, the data derived from electron-photon coincidence experiments<sup>12</sup> and the data of Mumma *et al.*<sup>13</sup> The other experimental data are limited to eight and six discrete energies, respectively, whereas our data are continuous over the complete energy range from threshold to 500 eV. We note the very good agreement with the Mumma *et al.* data at 50 eV and above. Their data at lower energies appear to be too low. We note also the good agreement

with the data of Steph and Golden except their 100-eV data point seems high. At lower energies, below 50 eV, their data are larger than ours due mainly to the depolarizing influence of cascade in our results. This is discussed further below. Perhaps surprisingly, the best agreement with theory is with the Born approximation calculation.<sup>25</sup> We note that this calculation is for  $2^1P$  excitation only. The predicted polarizations for  $3^1P$  excitation are slightly higher and thus if this fact was folded into the data the agreement between our data and the Born calculations

TABLE II. Parameters derived from the polarization data.

Target	Observed transition	$E_p$ (eV)	Calculated <sup>a</sup> $E_p$ (eV)	Estimated <sup>a</sup> ratio $\beta$ of cascade to direct excitation	$P_T$ [Eq. (5)]
He	$n^1P \rightarrow 1^1S$	$375 \pm 10$	440	0.05	0.99
Ne	various <sup>a</sup>	$320 \pm 10$	390	1.1	0.9

<sup>a</sup> See text for further details.

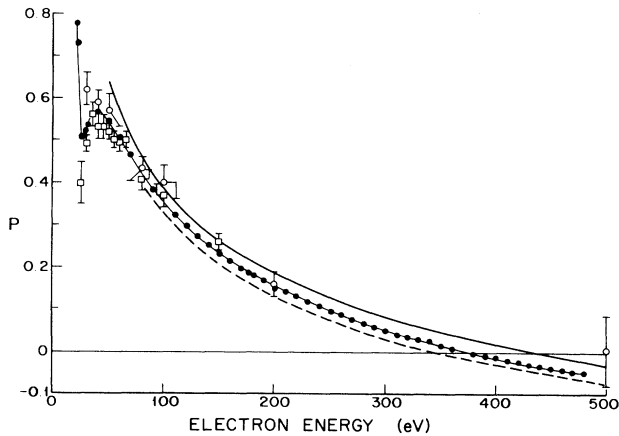


FIG. 1. Polarization of  $\text{He}(n\ ^1P-1\ ^1S)$  radiation as a function of electron-impact energy. ●, present data; ○, data obtained from electron-photon coincidence measurements (Ref. 12) of  $2\ ^1P$  excitation (see text); □, data of Mumma *et al.* (Ref. 13); ---, Born approximation (Ref. 25); —, Glauber approximation ( $3\ ^1P-2\ ^1S$  transition); data from first-order many-body theory (Ref. 28) are essentially identical to the Born data. For clarity error limits are not shown for our data, and data points are only given about every 10 eV. Error limits are given in Table I and are discussed in the text.

would be even closer.

If the cascade component of our observed signal is assumed to be unpolarized then it is possible to correct our measured data using the results of Donaldson *et al.*<sup>32</sup> and Westerveld *et al.*<sup>33</sup> for the fractional cascade components in order to obtain the polarization which would be obtained in the absence of cascade. This is then directly comparable with the Steph and Golden<sup>12</sup> data which are cascade free. It is easy to show that if the cascade fraction of the total measured cross section (assumed unpolarized) is  $f$  and if the measured polarization is  $P_M$  then the polarization, which would be obtained in the absence of cascade, is given by

$$P = \frac{3P_M(1+f)}{3+fP_M}. \quad (9)$$

Figure 2 shows our low-energy He data corrected in this way and compared with the Steph and Golden results. These are seen to lie between our two curves. We have assumed that all  $n\ ^1P \rightarrow 1\ ^1S$  radiation is similarly polarized, based on a review of existing data such as presented by Westerveld *et al.*<sup>33</sup> It is significant that both our cascade corrected data and the cascade-free data appear to "level out" towards lower energies. If no other processes were occurring we would expect the data to be extrapolating towards the threshold value of unity. Clearly this is not the case even in the energy region around 30 eV where no resonances occur. Some other mechanism, for example, the electron-electron correlation mechanism proposed by Heideman *et al.*,<sup>34</sup> must be responsible for this effect. Our energy resolution is inadequate to allow us to observe any resonance phenomena

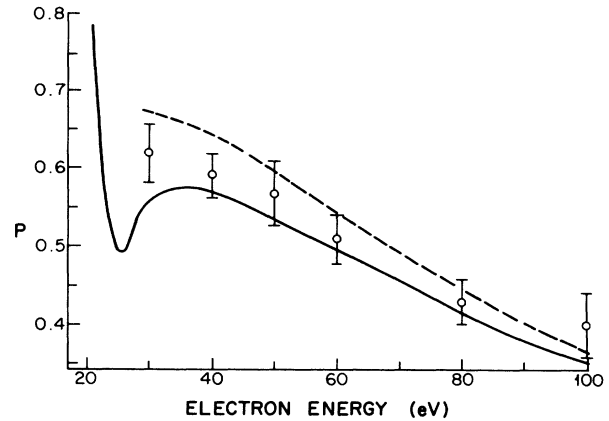


FIG. 2. Polarization of  $\text{He}(n\ ^1P-1\ ^1S)$  radiation as a function of electron energy. Solid curve, raw data. Dashed curve, data corrected for cascade contribution as discussed in text. Data points from electron-photon coincidence measurements (Ref. 12).

such as are known<sup>35</sup> to be a feature of the vuv emission from He in the near-threshold region. These will almost certainly be responsible for at least part of the sharp drop in  $P$  just above threshold.

The measured slope  $G'_A$  [Eq. (5a)] evaluated at  $P=0$  was  $-0.234$ . Assuming a value of  $\beta=0.05$ , which is consistent with the findings of Donaldson *et al.*,<sup>32</sup> leads to a value of  $P_T$  of 0.99. This coincides with the expected value of unity.

Values of  $C_j$  [Eq. (3a)] have been calculated and are listed by Donaldson *et al.* These range from 0.154 for the  $2\ ^1P$  state to 0.163 for the  $4\ ^1P$  state. Using the observed value of  $E_p$  for the integrated radiation we deduce a  $C_j$  value of 0.182 rather larger than any of the calculated values. This tendency for the Bethe approximation to overestimate the value of  $E_p$  has also been noted in the case of Lyman  $\alpha$  excitation in hydrogen.<sup>21</sup>

## B. Neon

The Ne data are shown in Fig. 3 and listed in Table I. The analysis of the Ne data is hampered by the fact that the resonance lines contain large cascade components<sup>36-38</sup> and also by the fact that the detector is sensitive to a wide spectral range. Thus the observed radiation may contain many spectral components. The main contributions will come from the resonance lines of the neutral atom at 73.6 and 74.4 nm and also those of the ion at 46.1 and 46.2 nm, though at the shorter wavelengths the quantum efficiency of our detector is considerably reduced.

It is clear from Fig. 3 that near-threshold resonance effects must be strong since the polarization collapses to zero in this region and shows no indication of a rise just at threshold as was seen with He. The polarization displays a broad maximum value of 0.39 at an electron energy of 38 eV before falling off to cross the energy axis at  $320 \pm 10$  eV. Relevant parameters are given in Table II.

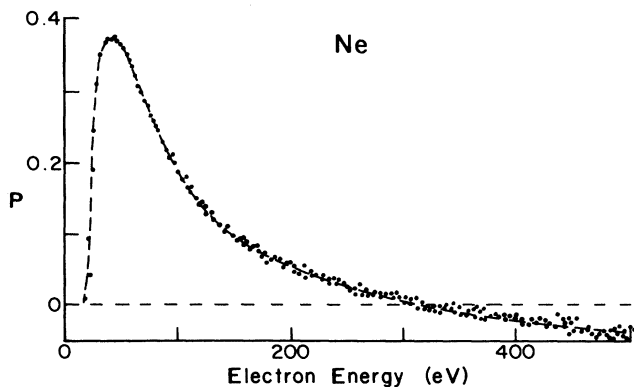


FIG. 3. Polarization of integrated vuv radiation from Ne as a function of incident electron energy (see text for further details).

Sharpton *et al.*<sup>38</sup> have made measurements of the main transitions which may contribute to the resonance radiation via cascade. They note that these transitions are unpolarized. We note also that the other main contribution to the observed radiation, the  $\text{Ne}^+$  resonance lines ( $2s2p^6\ ^2S_{1/2} \rightarrow 2s^22p^5\ ^2P_{1/2,3/2}$ ) at 46 nm, are also unpolarized. Thus we are basically observing the directly excited neutral resonance lines which are strongly polarized, but with a polarization which is reduced by cascade from higher states and by unpolarized contributions from other lines. If we consider all the unpolarized contributions as lumped together then we can evaluate an effective  $\beta$  parameter in Eq. (5a) and hence evaluate an approximate value for the threshold polarization  $P_T$ .

Phillips *et al.*<sup>37</sup> have measured the apparent cross section of the two neutral resonance lines to be  $7.8 \times 10^{-18}$  cm<sup>2</sup> at 300 eV with a cascade contribution of 29%. Assuming that other contributions are dominated by the two  $\text{Ne}^+$  resonance lines at 46 nm and allowing for the variation of our detection efficiency with wavelength we calculate an unpolarized background contribution at 300 eV equivalent to a cross section of  $3.9 \times 10^{-18}$  cm<sup>2</sup> (assuming Van Raan's<sup>39</sup> cross-section values). Thus we obtain an effective  $\beta$  [Eq. (5a)] of 1.1 and hence a  $P_T$  [Eq. (9)] of 0.9. This is very close (possibly fortuitously so given the uncertainties involved) to the value of unity which would be predicted for the resonance lines in the absence of cascade and other effects.

At 40 eV, which is below the threshold for the ion lines, the only depolarizing contribution will be from cascade. This has been measured for the 73.6- and 74.4-nm lines by Phillips *et al.*<sup>37</sup> They suggest a 36% cascade contribution at this energy. Assuming that this is unpolarized, we can predict a polarization for the resonance lines, at this energy, in the absence of cascade, of 0.57. This is rather close to the cascade-corrected polarization of the  $\text{He}(n\ ^1P - 1\ ^1S)$  emissions, Fig. 2.

To our knowledge there are no other measurements or calculations of Ne line polarizations with which we can compare our data. There have been some other measurements<sup>39</sup> of the parameter  $C_j$ , Eq. (3a). The most accurate of these is that of Van Raan.<sup>39</sup> Using his data together with Eq. (3a) leads to a value of  $E_p$  of 390 eV; this is somewhat higher than our measured value of  $320 \pm 10$  eV.

## VI. CONCLUSIONS

Accurate polarization data have been obtained for the integrated vuv radiation from He and Ne below 100 nm over the incident electron energy range from threshold to 500 eV. Resonance effects appear to be dominant in the near-threshold region, though in helium, the threshold polarization  $P_T$  was measured to be 0.78, a value consistent with unity given the energy spread in the electron beam. Cascade is observed to play a dominant role in Ne where very significant reductions in measured polarizations were due to this phenomenon. In He there is strong evidence for a further depolarizing effect, possibly involving a post-collision interaction of the type discussed by Heideman *et al.*,<sup>34</sup> in addition to cascade and resonance effects. An analysis based on the slope of the polarization curve in the energy region where it reversed sign led to predicted  $P_T$  values which were close to unity in both targets studied. This agrees with earlier work in the visible spectral region using different targets.

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\*Permanent address: Department of Physics, University of Manchester, United Kingdom.

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