Ratios of forward to back scattering of low-energy electrons by neutral gases*

R. C. Dehmel, M. A. Fineman,† and D. R. Miller
Department of Applied Mechanics and Engineering Sciences, and Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92037
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Measurements are reported on the ratio of forward to back scattering of electrons in the energy range 3–20 eV by He, O, Ar, H_2, N_2, and O_2 using the modulated crossed-beam technique. It is found that atomic oxygen has strong forward scattering at the lower energies. Agreement is found between the present results for He, Ar, H_2, and N_2 and ratios calculated from previously published differential-cross-section measurements except for H_2 and N_2 at the lower-energy limit.

I. INTRODUCTION

As atomic oxygen is the major constituent of the atmosphere between 200 and 700 km, many of the interesting phenomena, such as aurorae, involve electron-scattering processes by this atom. Despite this fact, no measurement has yet been reported of the differential scattering cross sections of low-energy electrons (<25 eV) by atomic oxygen; nor to date, has theoretical analysis of this open-shell atom led to differential-cross-section results, although encouraging calculations for total scattering processes have been reported.[1,2] Experimentally, two total-scattering cross-section measurements at low energies have been made[3,4] which agree within 30%. Recent calculations of electron-transport properties in the upper atmosphere[5] have suggested that an important parameter is the ratio of the total forward-to-back-scattered electrons at low energies. While such a ratio is obviously not as informative as complete differential data, it should provide an indication of the strength of polarization interactions which tend to give unsymmetrical scattering distributions even for s-wave scattering.

In this article, we report data for the intensity ratio of forward-to-back-scattered electrons which we have obtained for atomic oxygen and several other gases in the energy range 3–20 eV using the modulated-crossed-beam technique. While there are serious caveats in our simple method, a comparison with existing data for helium indicates that at least the qualitative energy behavior should be correct. Our data suggest an anomalously high degree of forward scattering of electrons by atomic oxygen below 7.5 eV.

II. METHOD, APPARATUS, AND UNCERTAINTIES

The experimental arrangement is shown in Fig. 1. Atomic oxygen is formed by dissociation of O_2 in an rf discharge, with subsequent expansion through a 0.05-cm nozzle at 15 torr. This nozzle source is a modified version[6] of an apparatus described previously.[7] The beam flux density is 10^{16}–10^{17} particles/(cm^2 sec) with 47% of the beam being atomic oxygen, as determined by a quadrupole mass spectrometer.[7] Because of the high densities in the nozzle expansion, our calculations have shown that, while only ground-state $O(^3P)$ and $O_2(^3\Sigma_g^-)$ are present in the final beam, some excited $O_2(^1\Delta_g)$ might survive due to the poor collisional deexcitation of this state.[6] While we have not been able to make a quantitative estimate of the fraction of $O_2(^1\Delta_g)$ with a mass spectrometer, previous data[8] on low-pressure rf discharges have shown fractions as high as 10%. We are now constructing a hexapole magnetic field to aid in a more quantitative determination of the beam composition for our system. The power in the discharge was reduced until the number of charged particles formed by the discharge was undetectable ($<10^{-13}$ A) as measured on the scattered-signal detection cups. For the other gases, the apparatus was run with the discharge off, as a simple nozzle source with a typical nozzle pressure of 25 torr.

The molecular beam was modulated at 206 Hz by a mechanical chopper wheel. The beam then passed through a 0.030-cm-wide slit skimmer into the second vacuum chamber, where the background pressure was 2×10^{-7} torr. A mask was placed downstream from the nozzle-skimmer arrangement to produce a 0.089-cm-wide beam at its point of intersection with the electron beam. After passage through the scattering region, the molecular beam was analyzed by a quadrupole mass spectrometer.

The electron gun consisted of standard Pierce-type electrodes for a cylindrical beam, followed by an asymmetric flat-plate einzel lens focusing element. The gun had a perveance of 0.03×10^{-6}

$A/V^{3/2}$, a current density of 0.25 mA/cm^2, and an energy spread of 0.25 ± 0.05 eV over the energy range as measured by the retarding-potential technique. An indirectly heated, oxide-coated cathode was used,[9] and all gun and detection parts were made of gold-blacked molybdenum.

To measure the forward- and back-scattered signals, the detection apparatus consisted of two signal detection cups facing one another each with a diameter of 3.18 cm, depth of 0.585 cm, and a 0.483-cm hole for passage of the electron beam, followed by an anode to collect the main electron beam. All electrons scattered into a forward or back conical angle of $20^\circ - 88^\circ$ from the scattering center were detected. The anode consisted of a 5.1-cm-long cylinder with a 0.635-cm entrance aperture and was held a few volts above ground. To prevent stray electrons, particularly from the gun, from reaching the detection cups, the gun and detection apparatus were enclosed in a stainless-steel can and separated from each other by an inner partition.

The scattered-signal detection cups were run at ground potential with the energy of the electrons defined by the potential difference of the detection cups and the cathode $-V_c$. The first and third einzel-lens elements (EL1, EL3) were at ground so that the electrons would be at the desired potential before entering the scattering region. EL2 was set at $(-V_c + 2.5\ \text{V})$. The apertures of EL1 and EL3 were 0.071 cm in diameter; that of EL2 was 0.142 cm. The distance of EL2 to EL1 and EL3 was 0.431 and 0.203 cm, respectively. An asymmetric lens of this type has been shown experimentally[10] to produce less spherical aberration than a symmetric lens with the same convergence.

Signals from each of the forward- and back-scattered-signal detection cups and the anode were impedance matched to the lock-in amplifier with 740 FET input operational amplifiers of input impedance $10^{12}\ \Omega$. The buffered signals were measured using a PAR HR-8 lock-in amplifier with a PAR-type B preamp. The input voltage to the 740's was developed across a 10-MΩ resistor. Since electrons impinging on the forward and back detection cups and the anode caused a negative potential, it was necessary to bias the low end of the resistor with a battery to return the detection cups to ground potential and the anode to a positive potential. The "measured" scattering ratio η_M is the ratio of the signals measured by the lock-in amplifier from the front and back detection cups.

In this type of experiment the question of electron reflection is important in determining the origin of the signals being measured; secondary electron emission is negligible compared with primary reflection at these energies. A consideration of the detection geometry shows that any background current on the back-scattered cup with the molecular beam off (flagged) must be due solely to reflection of that portion of the electron beam which falls on the forward-scattering region owing to space-charge spreading of the main beam; the amount of scattering from the residual background gases was negligible. This permits an *in situ* measurement of the reflection coefficient by measurement of the dc currents on the forward and back detection cups. The reflection coefficient ρ is given by $\rho = I_1/I_2$, where I_1 and I_2 are the back and forward cup currents, respectively; the same result is found even when multiple reflections are included in the calculation.[11] These coefficients, for electrons incident on gold-blacked molybdeum at greater than 20° to the surface normal, were found to be 0.07, 0.14, 0.21, 0.19, 0.27, and 0.34 for 3-, 5-, 7.5-, 10-, 15-, and 20-eV electrons, respectively.

In order to account accurately for reflection we would also need to know the reflection as a function of incident angle. Because the collectors are cups and not flat plates, a considerable fraction of the reflected electrons will strike a different section of the same cup, so that the effective reflection coefficient when molecular-beam scattering occurs will be decreased from our measured values. Thus the reflection coefficients given above, which we measured for 20° incidence, should represent a maximum reflection for our system, with the actual, but unknown, reflection being much lower. Assuming a constant effective reflection coefficient at each energy, the correct ratio η_{corr} is related to the measured ratio η_M by

$$\eta_{\text{corr}} = (\rho - \eta_M)/(\rho\eta_M - 1). \quad (1)$$

As discussed below, integration of available experimental differential cross sections over the

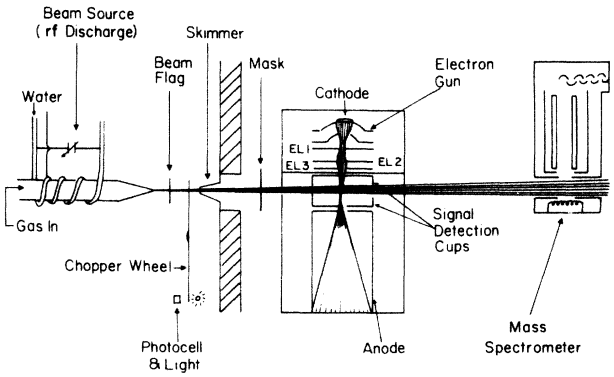

FIG. 1. Schematic diagram of the experimental apparatus (see text for dimensions).

angles we detected always gave ratios which lay between our measured ratios and our corrected (for reflection) results, but which were usually closer to the measured values.

To determine the scattering ratio for atomic oxygen, we use the formula

$$\eta(O) = \frac{y\eta(O+O_2)[1+\eta(O_2)][\sigma_T(O)/\sigma_T(O_2)] - [\eta(O_2) - \eta(O+O_2)]}{y[1+\eta(O_2)][\sigma_T(O)/\sigma_T(O_2)] + [\eta(O_2) - \eta(O+O_2)]}, \quad (2)$$

where $\eta(O)$, $\eta(O_2)$, and $\eta(O+O_2)$ are the appropriate forward- to back-scattered-signal ratios; y is (number density of O)/(number density of O_2 in the beam); and $\sigma_T(O)$ and $\sigma_T(O_2)$ are the total electron-scattering cross sections for O and O_2, respectively. This equation can be derived in a straightforward manner by considering the ratios of the forward- to back-scattered signals in terms of forward- and back-scattering cross sections and the number densities of the various species.

Equation (2) assumes that the sum of the forward- and back-scattered signals which were measured was equal to the total scattered signal. As signals scattered into 10% of the total sphere were not detected, this assumption involves some error. However, this error is very small since we are only concerned with the ratio of total signals [written in terms of the total cross sections $\sigma_T(O)/\sigma_T(O_2)$], and as this ratio is close to unity (~0.8–1.0), the effect of the signals scattered into the unmeasured conical angles (0–20°, 88°–92°, and 160°–180°) tends to cancel. Without knowledge of the exact concentration of $O_2(^1\Delta_g)$, nor its total cross section, we have assumed that $\sigma_T(O_2)$ is that due only to ground-state O_2. We have used the values given by Sunshine *et al.*[4] for $\sigma_T(O)/\sigma_T(O_2)$; extrapolation was necessary above 11.3 eV.

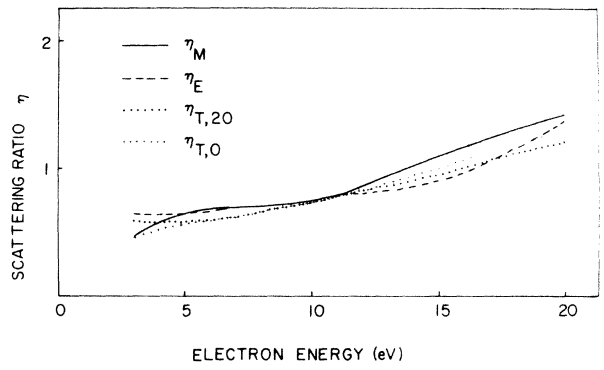

FIG. 2. Ratios of forward to back scattering of electrons by He as a function of electron energy. The solid line, η_M, represents our measurements; the dashed line, η_E, is computed from integration of the experimental angular results of Gibson (Ref. 8); the dotted line, $\eta_{T,20}$, and the crossed line, $\eta_{T,0}$, are computed (see text) from the theoretical results of LeBahn (Ref. 13).

III. RESULTS AND DISCUSSION

Experimentally and theoretically, helium is the most widely studied gas. In addition, the experiments of Gibson and Dolder[12] on helium are probably the most definitive angular measurements made to date. We have therefore made a careful comparison of our results for helium with those of Gibson and with the theoretical results of LaBahn and Callaway[13] which is shown in Fig. 2. Here η_M is our measured forward- to back-scattering ratio, η_E is the ratio obtained from the angular measurements of Gibson and Dolder integrated from 20° to 90° and from 90° to 160°; $\eta_{T,20}$ is the ratio obtained from the theoretical results of LaBahn integrated from 20° to 90° and from 90° to 160°; and $\eta_{T,0}$ is the ratio obtained from LaBahn's results integrated from 0° to 90° and from 90° to 180° to see how significantly the ratios are affected by the missing solid angles in the detection apparatus. It should be noted that these ratios, whether obtained by experiment directly or indirectly (η_M or η_E), or theoretically (η_T), show similar values and similar trend with electron energy.

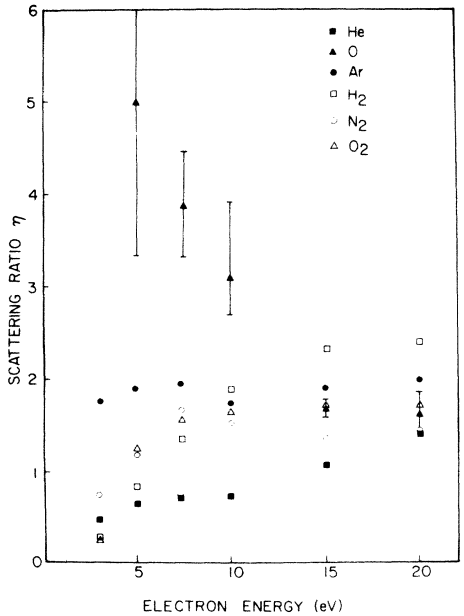

FIG. 3. Experimental ratios of forward to back scattering of electrons by He, O, Ar, H_2, N_2, and O_2.

For Ar, H_2, and N_2, the integration of angular data, although not shown here, gives ratios which lie roughly 10% above our measured values except for H_2 and N_2 at the outer energy limits where the integrated angular data show much stronger forward scattering than our results do. The experimental differential scattering cross sections used for comparison were those of Ramsauer and Kollath[14] for Ar, and Bullard and Massey[15] for H_2, and Massey and Bullard[16] and Shyn *et al.*[17] for N_2.

Because of this agreement with other data, and for the reasons discussed above with regard to effective reflection coefficients, we show our results, for all gases studied, in Fig. 3 without corrections for electron reflection. These results should then form a lower limit on the actual ratio if $\eta_M > 1$ and an upper limit on the ratio if $\eta_M < 1$; see Eq. (1).

The error bars shown for the atomic oxygen are based on our experimental reproducibility and not on uncertainties in Sunshine's data. Data at 3 eV are not reported for atomic oxygen since the errors were too large. However, the mean of the 3-eV data did suggest that the ratio decreases from the 5-eV data. Omitting the error for reflection correction, errors for all other gases are estimated to be less than 20% below 7.5 eV and less than 15% at higher energies. The large uncertainty in the values for atomic oxygen can be traced to poor experimental signal to noise magnified through Eq. (2). Because of the rf discharge properties, the atomic-oxygen-beam density was lower than in the other cases by a factor of at least 5. In addition, continuous operation with atomic oxygen apparently inhibits cathode emission, giving generally lower electron-beam currents.

Unfortunately, as previously discussed, we cannot assess the effect of $O_2(^1\Delta_g)$ on the values we have obtained. Even if $O_2(^1\Delta_g)$ contributed as much as 10% to the scattering process, Eq. (2) shows that it would require a very large cross section, with very strong forward scattering ($\eta_M > 10$), to negate the qualitative conclusion that atomic oxygen shows strong forward scattering below 10 eV.

As we mentioned above, consideration of reflection coefficients will only yield larger values of η_M for atomic oxygen. Also, if we use the earlier data of Neynaber *et al.*[3] for $\sigma_T(O_2)$ instead of Sunshine *et al.*,[4] the values of η_M for atomic oxygen are again slightly higher. We do not presently have a theoretical explanation for the strong forward scattering by atomic oxygen at low energies. It is clear that polarization effects will be important and that with low-lying excited states ^1D and ^1S, strong coupling between states must be considered.

For the other gases we find nothing unexpected except for H_2 and N_2 below 5 eV. These data were reproducible, especially for H_2, and suggest that there is stronger backscattering than the differential data have indicated.[15,16]

It is clear from these simple ratio measurements that an interesting variety of low-energy behavior exists for atmospheric species. A review of the literature also indicates gaps in low-energy differential electron-scattering data, especially below 7.5 eV. We hope that the present data will encourage additional experiments in this energy range.

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†Permanent address: Department of Physics, Lycoming College, Williamsport, Pa.

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