the Ramsauer-Townsend minimum in the momentumtransfer cross section. Hence, the total cross section in the neighborhood of the minimum is given by the  $P$ -wave phase shift while the momentum-transfer cross section is approximately given by the difference between the P- and D-wave phase shifts. Therefore, one would expect the minimum momentum-transfer cross section to lie deeper than the minimum total cross section. The fact that the results of Frost and Phelps<sup>16</sup> did not find such a deep Ramsauer-Townsend minimum must be attributed to their necessarily broad distribution of electron energies.

In the case of argon, where there is a strong energy dependence of the cross section, it is quite clear that a much better measure of the momentum-transfer cross section is obtained from precise measurements of total cross sections than from the unfolding of swarmexperiment data.

In the case of argon, the cross section is a rapidly varying function of electron energy and hence it is more difficult to evaluate the precision in the determination of the effective-range parameters. It can be stated that A and B are less precise, and  $A_1$  more precise than is the case in helium. Computations made by varying the parameters using the same criterion as used for helium showed that a change of  $\pm 3\%$  in A and  $\pm 5\%$  in eithe B or  $A_1$  will be detected.<sup>25</sup> B or  $A_1$  will be detected.<sup>25</sup>

<sup>25</sup> O'Malley in Ref. 2 has estimated a precision of about  $2\%$  in the determination of A for argon from the data of Ramsauer and Kollath.

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# Scattering Cross Sections of Argon and Atomic Oxygen to Thermal Electrons\*

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The electron-scattering cross section for momentum transfer of several neutral species has been experimentally determined. An S-band microwave interferometer was employed to investigate the plasma produced by the incident shock wave in a shock tube. This plasma was in the state of equilibrium, so the relative species concentrations and the electron velocity distribution were known. For electron-collision frequencies small compared with the signal frequency, the microwave data could be intepreted to yield the scattering cross sections. The cross section of the argon atom was measured from 1800 to 5500'K. The results agree with previous drift measurements when those cross sections are averaged over the Maxwellian distribution of electron velocities. Above 3000'K the oxygen molecule becomes dissociated and the oxygen-atom cross section can be measured. From 3000 to 4000°K, this atomic cross section is  $(1.2\pm 0.2) \times 10^{-15}$  cm<sup>2</sup>. For molecular nitrogen and oxygen the small-collision-frequency requirement could not be fully satisfied. The cross sections for these gases were approximately  $10^{-15}$  cm<sup>2</sup> from 2000 to 3500°K, with no observable dependency on gas density.

## INTRODUCTION

EASUREMENTS have been made in the past of  $\blacktriangle$  the electron-scattering cross section in argon<sup>1</sup> and molecular nitrogen<sup>2,3</sup> using a swarm technique. The molecular oxygen-scattering cross section has been measured up to 900'K by Mentzoni4 using a microwave interferometer. The atomic-oxygen cross section was measured at 4000°K by Lin,<sup>5</sup> using a combination of microwave-attenuation and conductivity probe data. To extend the data range of the atomic-oxygen species and to obtain data for argon in a similar environment, the present shock-tube experiments were performed.

The plasmas are produced by a shock wave in a shock tube. This plasma generator was selected because the species distribution can be calculated from the equilibrium normal shock-wave relations. The operating conditions were selected so that the electrons in the shock tube would be thermally equilibrated with the neutrals; therefore, their temperature can be taken to be the same as the temperature of the neutrals which was varied from 2000 to 5000'K. The cross sections which are obtained are then averages over a Maxwellian velocity distribution. By taking the ratio of the measured attenuation and phase shift of an S-band microwave signal propagating through the plasma, the electron collision frequency can be determined. From this collision frequency the scattering cross sections to electrons could be inferred because the species distribution and electron temperature were known.

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## EXPERIMENTAL CONSIDERATIONS

In the present experiment, microwaves were propagated through the shock tube as they would be in an ordinary waveguide. The shock wave traveled in the oppositve direction to the microwaves. The shock wave processed the low-pressure, room-temperature test gas to a high temperature and pressure. The operating conditions were selected so that the time required for the gas to relax to equilibrium would be rapid. The depth of the processed plasma extends from the shock wave to the gas interface separating the plasma from the lowtemperature helium driver. As the shock wave advanced down the shock tube, the plasma depth continually increased at a rate equal to the difference between the particle velocity behind the shock wave and the shock-wave velocity. Consequently, the amounts of attenuation and phase shift experienced by the microwaves from their interaction with the plasma were also continually increased.

The 3000-Mc microwaves were introduced through a quartz window, (see Fig. 1) which formed the end wall of the  $1\frac{1}{2}$ -in.  $\times 2\frac{1}{2}$ -in. shock tube. The microwaves were removed from the shock tube just before the driver section by a coupler which had good directivity. The phase change in the test signal which propagated through the plasma was detected, using an amplitude-insensitive phase-detecting network. '

### Data Reduction

Since the microwaves are always propagating in a waveguide the assumption can be made that plane waves are interacting with the plasma. Measurements made on dielectric samples of known properties which were placed in the shock tube gave the correct result within  $\pm 10\%$  when plane-wave theory was used to reduce the data.

From this plane-wave theory, the ratio of the microwave attenuation A (in decibels) to the phase shift  $\beta$  (in



FIG. 1. Schematic diagram of the S-band microwave interferometer.

<sup>8</sup> R. J. Blum, Proc. IEEE 53, 523 (1965).

degrees) of a low-power, plane microwave signal traversing an under-dense uniform plasma slab  $(\nu_n^2 \ll \omega^2)$ is given by the approximate relation'

$$
A/\beta = (20\pi/180 \ln 10)(\nu/\omega)(1 - \nu_p^2/4\omega^2 \cos^2 \varphi),
$$

where  $\omega$  is the circular frequency of the incident microwave beam,  $\varphi$  is its angle of incidence, and  $\nu_p$  is the electron plasma frequency. There are two features of this relationship which are important for the present shock-tube studies; namely the ratio  $A/\beta$  is only weakly dependent on the electron density and is independent of the plasma thickness. Consequently, from the measured ratio of attenuation to phase shift, the electron collision frequency  $\nu$  can be calculated.

In the derivation of this relation a uniform plasma with a constant electron density was assumed; however, in the shock-produced plasma there is an axial variation in the electron concentration. This distribution is a result of the finite time required for electron production behind the shock wave and of mixing at the driverdriven interface.

Previous calculations of the effect of electron density gradients at plasma boundaries on electromagnetic signals<sup>8,9</sup> have shown that if the plasma is lossy and if the extent of the gradient region is small compared to a wavelength, then the particular distribution used for calculating transmitted phase and amplitude is not important. This same result also applies to the finite thickness plasmas produced in the shock tube. The data in Ref. 9 can be used for a trapezoidal distribution of electron density. Maintaining the extent of the gradient regions as constant and varying only the constant portion of the trapezoid, a comparison can be drawn with the changes induced by this variation to that of a uniform plasma of the same over-all depth. The results for plasma typical of those in the experiments showed a constant difference in phase and attenuation between the two distributions as a function of plasma depth. Consequently, since the plasma depth is uniformly increasing with time

$$
\left(\frac{\partial A}{\partial t}\middle/\frac{\partial \beta}{\partial t}\right)_{\text{trapezoid}} = \left(\frac{\partial A}{\partial t}\middle/\frac{\partial \beta}{\partial t}\right)_{\text{uniform plasma}}.
$$

Thus low-power, plane-wave theory for microwaves interacting with a uniform-plasma slug can be used to analyze the shock-tube data.

The quantities recorded on the oscilloscopes were the attenuation  $A$  and phase shift  $\beta$ . The raw data were reduced to decibels and degrees by adjusting the precision attenuator and phase shifter shown in Fig. 1, so

<sup>&</sup>lt;sup>7</sup> J. A. Stratton, *Electromagnetic Theory* (McGraw-Hill Book Company, Inc., New York, 1941), p. 500.<br><sup>8</sup> J. W. Daiber and H. S. Glick, in *Proceedings of the Symposiu* 

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that a point-by-point reproduction of the original traces was obtained. These plots then should have shown a linear increase of these quantities with time. Within the first  $150 \mu$ sec of laboratory time, the quantities increased quite slowly; this was probably due to relaxation effects. After this initial period, they increased rapidly and linearly with time. A straight line could be fitted to these data and the slope computed with almost no ambiguity. For some runs a slight ripple did exist in the data. This could be removed from the next run by readjustment of the slide screw tuners incorporated into the interferometer for the reduction of mismatches.

## Scattering Cross Section

The collision frequency can be related to a scattering cross section by using the relation

$$
v = \bar{c} \sum_i n_i Q_i(T_e) ,
$$

where  $n_i$  is the number density of the *i*th species which has a cross section  $Q_i(T_e)$ , and the mean electron speed is given by

$$
\bar{c} = (4/3)(8kT_e/\pi m)^{1/2}
$$

which depends on the electron temperature  $T_e$ .

Margenau<sup>10</sup> has derived an expression for the complex plasma conductivity when the electrons have a Maxwellian velocity distribution and are subjected to highfrequency electromagnetic 6elds. By using the usual microwave-plasma formula for the conductivity" and equating the real parts of these two equations, Margenau's result can be used to define an effective collision frequency.

When the simple kinetic theory expression for the collision frequency is equated to the effective collision frequency and the assumption is made that the collision frequency is much less than the signal frequency  $(\nu^2 \ll \omega^2)$ , then one obtains an expression for the averaged scattering cross section of the *i*th species:

$$
Q_i(T_e) = \frac{1}{2} \int_0^\infty Q_i(\epsilon) \left(\frac{\epsilon}{kT_e}\right)^2 \exp\left[-\frac{\epsilon}{kT_e}\right] \frac{d\epsilon}{kT_e}.
$$

The quantity  $Q_i(\epsilon)$  is the momentum-transfer cross section for a monoenergetic beam of electrons in which  $\epsilon$  is the energy of a single electron. These monoenergetic cross sections are given as a function of energy, usually in the units of electron volts; however, by dividing the energy by the Boltzmann constant, these monoenergetic cross sections can be plotted on a temperature scale. Since the integral extends over all electron energies, the value of  $Q(T_e)$  can be quite different from the corresponding value of  $Q(\epsilon)$  where  $\epsilon/k = T_{\epsilon}$ . This difference is especially striking for argon in the vicinity of the Ramsauer minimum where the high-energy portion of



FIG. 2. Scattering cross section of argon.

the distribution can make  $Q(T_e)$  much greater than  $Q(\epsilon)$ ; for example, at an electron temperature of 4000°K this difference is approximately a factor of 10 (see Fig. 2).

If the present data had been obtained over a wide temperature range, then the values of  $Q(\epsilon)$  could be found directly. This would be done by finding that variation of  $Q(\epsilon)$  which would best predict our measurements. Since the temperature range was rather limited, a comparison with previous data was made by averaging that earlier data to obtain  $Q(T_e)$ .

For electron densities comparable to or above the critical value of  $4.26\times10^{10}$  cm<sup>-3</sup> the microwave attenuation ation was too large to be determined. This controlled the maximum temperature for a given gas density at which data could be obtained. The gas density could be lowered until low-pressure shock tube effects<sup>12</sup> reduced the testing time to less than the times required for relaxation to equilibrium. The upper limit on gas density was established by the requirement that the collision frequency be much less than the signal frequency. For all of the runs shown in the figures, the ratio of  $\nu^2/\omega^2$ was in the range from 0.01 to 0.1.

### Species Equilibrium

The interpretation of the collision frequencies inferred from the microwave measurements as scattering cross section requires that the concentration of all plasma species  $n_i$  be known. When there is dissociation, the equilibrium distribution of species can be calculated from the initial gas composition and pressure and the shock-wave velocity. To verify that equilibrium is achieved in the present shock-tube experiments, a numerical calculation of the flow behind a normal shock. wave (using finite rate chemistry) was performed. The reaction rates used were taken from Ref. 13. It was found that the time required for the oxygen-atom con-

<sup>&</sup>lt;sup>10</sup> H. Margenau, Phys. Rev. 69, 508 (1946).<br><sup>11</sup> M. A. Heald and C. B. Wharton, *Plasma Diagnostics with*<br>*Microwaves* (John Wiley & Sons, Inc., New York, 1965), p. 6.

<sup>&</sup>lt;sup>12</sup> R. E. Duff, Phys. Fluids 2, 207 (1959).<br><sup>13</sup> A. Q. Eschenroeder, J. W. Daiber, T. C. Golian, and A. Hertzberg, *High Temperature Aspects of Hypersonic Flow* (Per-<br>gamon Press, Inc., New York, 1963), p. 27. gamon Press, Inc., New York, 1963), p. 27.

centration to reach equilibrium in  $5\%$  O<sub>2</sub>+95% Ar mixture at 3 Torr initial pressure in the shock tube is 30  $\mu$ sec when the final temperature is 3500°K. This relaxation time is to be compared with the total estimated test time of 400  $\mu$ sec.

The time required for ionization to reach equilibrium in this pure plasma is over  $3000 \mu$ sec. Thus, the observed degree of ionization cannot be due to ionization of the pure gas species. This implies that the electrons forming the plasma originate from impurities. The ionization energy for the impurities is low which suggests that the ionization may be rapid and that equilibrium may be quickly achieved.

These impurities do not affect the measured electron collision frequency in the underdense plasma, since the product of number density and collision cross section of the impurities is much smaller than that of the constituent gas. The observed ionization level at 3500'K could be accounted for by a sodium impurity concentration of only four parts in 10". This level of impurity concentration would certainly be expected in the shock tube.

### Energy Equilibrium

The temperature of the electrons must also be known, not only to assign the correct temperature to the cross sections, but also to determine the mean electron velocity. The gas is heated behind the shock wave to high temperatures. For most of the present cases, the translational temperature of the neutrals is within  $3\%$ of the equilibrium temperature after 30  $\mu$ sec. This equilibrium temperature which is easily calculated from the shock-wave conservation equations is then maintained throughout the plasma slug. If the free electrons could exchange energy with the neutrals fast enough, then they would also be maintained at this equilibrium temperature.

For the argon plasmas, an electron-energy relaxation time can be computed using the hard-sphere energytime can be computed using the hard-sphere energy<br>exchange rate.<sup>14</sup> For the initial shock-tube pressure used (100 to 15 Torr), these times varied from 8 to 80  $\mu$ sec, which is within the available testing time. The experiments of Schulz<sup>15</sup> have indicated that the vibrational energy states of molecules have a large cross section to electrons. It thus seems reasonable to assume that the before dissociation has occurred the electronenergy distribution will be closely coupled to the vibra-<br>tional-energy distribution.<sup>16</sup> For the  $5\%$  O<sub>2</sub>+95% A tional-energy distribution.<sup>16</sup> For the 5%  $O_2+95\%$  Ar mixtures, the time required for vibrational energy to relax to the translational energy<sup>17</sup> was found to be 25  $\mu$ sec at 3500°K for an initial shock-tube pressure of 2 Torr. The actual rate will be still faster because of the elastic contribution. It can thus be concluded that for the plasmas of interest an energy equilibrium between the electrons and the plasma is soon reached. Once this equilibrium has been achieved, the electron-temperature distribution through plasma slug remains constant to the interface. If a temperature equilibrium had not been achieved in the plasmas of these experiments, then the ratio  $A/\beta$  would have changed as the electron temperature changed. However, the observed variations of A and  $\beta$  were completely linear with time and consequently the ratio  $A/\beta$  was constant.

#### SCATTERING CROSS-SECTION RESULTS

## Argon

For the pure argon plasma, the summation over species and cross section for the determination of collision'frequency becomes a single term. The measured attenutation and phase shift can then be immediately reduced to a collision cross section. The final results for argon are shown in Fig. 2. The cross section increases with temperature with no indication of a minimum. The variation of monoenergetic momentum cross section used by Frost and Phelps' to explain their drift experiment data is shown as the dot-dash line. The electron energy has been divided by the Boltzmann constant to obtain a temperature unit for their monoenergetic electrons. The cross sections of Frost and Phelps when numerically integrated are shown by the solid line, which is in fair agreement with the present data. There appears, however, to be a  $30\%$  displacement between the two sets of data. For the reduction of the data taken in argon-oxygen mixtures, the cross section which was used for the argon is given by the dashed line in Fig. 2.

## Atomic Oxygen

The data obtained from the  $5\%$  O<sub>2</sub>+95% Ar mixtures in the 3300- to 3800'K temperature range were used to determine the oxygen-atom cross section. The equilibrium values of the ratio of atomic to molecular oxygen concentration for these runs varied from 3 to 100, indicating that a small oxygen molecule contribution did exist for some runs. The correction of the data for the contribution of argon never exceeded 50% and that of  $O<sub>2</sub>$  never exceeded 10%. These data shown in Fig. 3 were obtained at shock wave speeds in the vicinity of  $2.2 \times 10^5$  cm/sec with driver tube pressures from 1.5 to 3 Torr.

The only other measurement of the atomic-oxygen cross section in the thermal-energy range is that by Lin and Kivel.<sup>5</sup> They deduced their point from measurements made with a conductivity probe and microwave attenuation data. Their value, shown in Fig. 3 as the circle, is lower than the present data.

Several of the available theoretical estimates of the atomic-oxygen cross section are shown in Fig. 3. One

<sup>&</sup>lt;sup>14</sup> J. M. Anderson and J. Goldstein, Phys. Rev. 102, 933 (1956).<br><sup>15</sup> G. J. Schulz, Phys. Rev. 125, 229 (1962); also, G. J. Schulz<br>and J. T. Dowell, *ibid.* 128, 174 (1963).<br><sup>16</sup> I. R. Hurle, J. Chem. Phys. 41, 3592 (196

of the earliest was that by Mitra, Ray, and Ghosh.<sup>18</sup> A more recent estimate based on a comparison with the measured photodetachment cross section of oxygen has measured photodetachment cross section of oxygen ha<br>been made by Klein and Brueckner.<sup>19</sup> Our experiment: data appear to agree with this curve. The scattering curves predicted by Bauer and Browne" and Bates and  $Massev<sup>21</sup>$  are lower than our data, but in reasonable agreement with Lin's datum point. The most recent prediction of the atomic-oxygen cross section is that of Garrett and Jackson.<sup>22</sup>

## Molecular Nitrogen and Oxygen

The same technique as described before was also applied to molecular oxygen and nitrogen plasmas. These gases have a larger cross section to electrons than does argon. Consequently, the ratio of  $v^2/\omega^2$  for our operating pressures was 0.2 to 5. The data reduction procedure outlined above is thus no longer valid. However, it can still be formally applied. The cross sections are then still be formally applied. The cross sections are ther<br>approximately 10<sup>-15</sup> cm<sup>2</sup> for electron temperatures from 2000 to 3500'K. This agrees with the nitrogen data of Engelhardt, Phelps, and Risk' and with a square-root temperature extrapolation of the molecular-oxygen data of Mentzoni.<sup>4</sup> The initial shock-tube pressure of both the nitrogen and the oxygen was varied from 1 to 10 Torr with no systematic dependency being observed for the variation of cross section with density.

- <sup>18</sup> S. K. Mitra, B. B. Ray, and S. P. Ghosh, Nature 45, 1017  $(1940)$ .
- $\frac{10 \text{ M}}{1958}$ . M. Klein and K. A. Brueckner, Phys. Rev. 111, 1115 (1958). <sup>20</sup> E. Bauer and H. N. Browne, in Atomic Collision Processes,
- edited by M. R. C. McDowell (Interscience Publishers, Inc. , New
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<sup>21</sup> D. R. Bates and H. S. W. Massey, Proc. Roy. Soc. (London)<br> **A192**, 1 (1947).
- W. R. Garrett and H. T. Jackson, Jr., Bull. Am. Phys. Soc. 11, <sup>495</sup> (1966); Army Missile Command Report RN-TR—66-4, 1966 (unpublished).



FIG. 3. Scattering cross section of atomic oxygen.

Runs were also made with oxygen diluted in argon. In this manner, the collision frequency could be lowered without decreasing the initial pressure. Several runs were made for which  $v^2/\omega^2$  was the order of 0.1. These molecular oxygen cross sections were also  $10^{-15}$  cm<sup>2</sup>, the molecular oxygen cross sections were also  $10^{-15}$  cm², the same as the higher pressure results.

### CONCLUSIONS

From the present shock, tube experiments, the elastic scattering cross section of argon to thermal electrons was obtained and compared to that of Frost and Phelps; the values are within  $30\%$  of each other. The cross section of atomic oxygen was obtained at 3500°K. The value was higher than that of Lin and Kivel and agreed with Klein and Brueckner's theoretical prediction. For the molecular gases  $N_2$  and  $O_2$ , the lowcollision-frequency requirement was not fully satisfied, but formal application of the data-reduction procedure but formal application of the c<br>gave cross sections of  $10^{-15}$  cm<sup>'</sup>