

THE OPTICAL EXCITATION FUNCTION OF HELIUM

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(Received September 6, 1930)

ABSTRACT

With a discharge tube which has been considerably modified from the designs previously used, and with a technique specially developed to eliminate the effects of collisions of the second kind and ionization and recombination, the author has completed a study of the excitation function of helium for electrons with energies between the excitation potentials and one hundred volts. Two new features are introduced, namely, experimental conditions under which a linear dependence of intensity on either current or pressure is obtained, and an actual determination of the electron velocity distribution curve. The intensity curves, by themselves, agree fairly well with those obtained by earlier workers but, by the innovations mentioned above, it has been possible to correct these curves to obtain the true optical excitation function values. The corrected results show that each line has a maximum excitation probability either at or within a few tenths of a volt of the excitation potential, and that the probability drops off quite rapidly above this value, more rapidly for the triplet system than for the singlet.

INTRODUCTION

SINCE the publication of the original work of Hughes and Lowe¹ on the variation of the intensities of the lines of the helium spectrum with change of velocity of the exciting electrons, a large number of workers have made studies in this field. While the results of these investigations have been somewhat discordant, two papers, those of Hanle² and Elenbaas,³ give results which show at least qualitative agreement between themselves and with the publication mentioned above. A study of the intensity-voltage curves given by these authors seems to enable certain conclusions to be drawn:

(1) All lines of the triplet system appear at their excitation potentials and show sharp maxima somewhere between these potentials and 35 volts.

(2) All lines of the singlet system increase in intensity rather rapidly just above the excitation potential, and show moderately flat excitation curves, with maxima between 60 and 150 volts.

While these points have been fairly definitely established in earlier investigations, the author has felt that certain discrepancies in the published results demanded further investigation. Elenbaas, for example, finds double maxima for most of the lines while Hanle, Hughes and Lowe find but one maximum for each line. The positions of the maxima also vary quite widely among the different investigations.

Before entering into a discussion of the present work, it may be well to

¹ Hughes and Lowe, Proc. Roy. Soc. **A104**, 480 (1923).

² Hanle, Zeits. f. Physik **56**, 94 (1929).

³ Elenbaas, Zeits. f. Physik **59**, 289 (1930).

define just what interpretation shall be placed on the term "optical excitation function." In order that spurious results due to the apparatus may be eliminated, it would seem well to define this function as *the relative probability that light of a particular wave-length be emitted when an isolated atom experiences a collision (defined, for example, by kinetic theory considerations) with an electron moving with a specified velocity.* This definition imposes two definite restrictions on the experimental conditions:

(1) The requirement of isolation demands that the effects of collisions of the second kind, collisions with the walls, etc., be kept negligible, and that there shall be not more than a negligible amount of light emitted due to ionization and subsequent recombination.

(2) The exciting electron beam must be as nearly homogenous as possible, and the extent of departure from homogeneity must be determined.

The work now being reported was undertaken with these limitations definitely in mind and an attempt has been made to devise tests showing to what extent each requirement has been satisfied.

THE DISCHARGE TUBE

The final design of discharge tube used for the excitation of the spectrum is shown in Fig. 1. Electrons from the ten mil tungsten filament were accelerated through the first slit, 2 by 20 mm, which was about 2 mm distant from

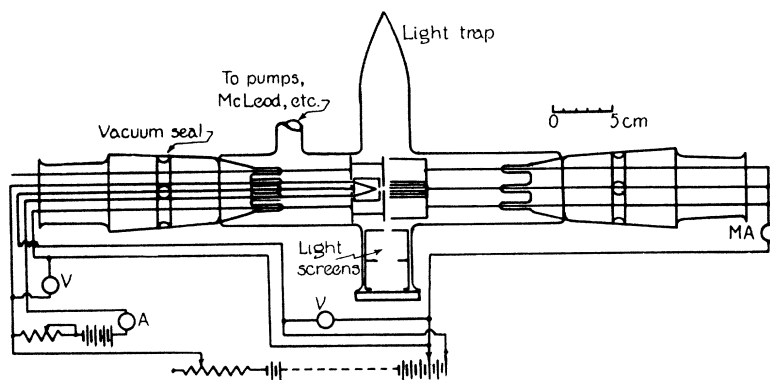


Fig. 1. Discharge tube with wiring diagram.

the filament. This slit was maintained at a potential difference of about sixteen volts above the second slit, about one mm away, and of the same size. Beyond this was a clear, field free space of 4 mm length, in which the emission was observed, and the plate, which was composed of a number of parallelo-piped boxes, 20 mm deep. All electrodes were of nickel. The various potentials were maintained by a bank of storage cells.

A remark in regard to the choice of the filament may be desirable. In the early part of the work tubes with oxide coated platinum emitters, usually of the unipotential type, were tried, in the hope of obtaining a very homogenous electron beam. Velocity distribution curves taken on these tubes showed,

invariably, a very large percentage of slow electrons, distributed between zero velocity and that applied to the slits. This difficulty was finally traced to secondary electrons, emitted from the slits, due to barium distilled onto their surfaces from the cathode. With its low work function, this material acted as a particularly efficient secondary emitter. The loss in homogeneity due to the voltage drop across the tungsten filament was much more than compensated for by the decrease in these secondaries.

In addition to the change to a tungsten filament, certain other precautions against secondary electrons were taken. The difference of potential, previously mentioned, between the two slits served to sweep back any secondaries formed at the edges of the second slit. The particular plate design used was also adopted for the elimination of this effect. With the large metal surface and the small opening obtained by this construction, the plate acted as a black cavity, not only for the electrons but also for ultraviolet light from the excited atoms. Reflected, secondary, and photo-electrons were therefore neither recorded in the current measurements nor allowed to enter the space in which light emission was observed.

The tungsten filament introduced a further difficulty in the large amount of continuous light which it furnished. This light gave a background on the plates which was comparable with the exposures produced by the lines themselves until the light trap and screens shown were introduced. The tube was painted, outside, with a dead black lacquer, with the exception of the plate glass window through which observations were taken.

Frequent filament replacements necessitated the ground glass joint construction shown in the figure. Stop-cock grease was used only on the outer portions of the joints, and the vacuum seals were pumped continuously except during the actual exposures, to keep vapor from reaching the electrodes. The construction prevented baking, however, and the following method was used for cleaning the electrodes. After the apparatus had been opened to air, a charcoal trap, in close proximity to the tube, was baked at 350–400°C for about twelve hours. Liquid air was then placed on this trap and the filament emission gradually built up until about four milliamperes flowed to the plate, with an accelerating potential of 300 volts. Under these conditions the electrodes, near the filament, were glowing at a dull red. During the clean-up action, if the filament current was held constant, the current to the plate increased for a few hours, then decreased slightly to a constant value. In general, the heating and bombardment were continued for ten to twelve hours, with the vacuum pumps running. The tube was then ready for operation. For subsequent exposures, if air had not been admitted, bombardments of three to four hours were found to be sufficient. This clean-up technique was found to be absolutely essential, as will be explained later.

The current vs. retarding potential curve shown in Fig. 2a was obtained with this tube after such treatment and with the pumps maintaining a vacuum of about 10^{-5} mm as measured by a McLeod gauge. The accelerating potential was 100 volts. The graphical differentiation of this curve gives the velocity distribution curve of Fig. 2b. By far the greater part of the elec-

tron beam is included within a five volt band, just accounted for by the filament potential drop. As the retarding potential was applied between the last slit and the plate, both of nickel, the 1.2 volt difference between the fastest electrons and the accelerating potential may be taken as the contact potential. This measurement checked, within the accuracy to which the voltage could be held constant during an exposure (about ± 0.3 volt), with the first appearance of the various lines at their excitation potentials. The "tailing off" of the distribution curve toward lower velocities may probably be attributed to a certain amount of electron reflection and secondary emission

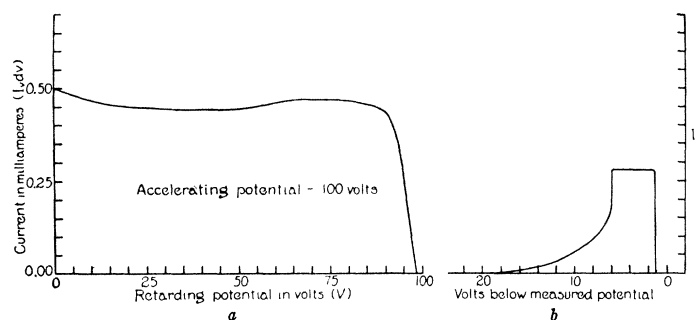


Fig. 2. Electron velocity distribution.

which was not eliminated. This region, however, accounts for only 30 percent of the total current, and this is much better than was obtained with any other type of tube construction which was tried.

The helium was purified as previously described⁴ and was passed through the charcoal trap before entering the tube. This helium was never left in the tube for more than three or four hours of operation, after which it was pumped out and the clean-up process was repeated, before fresh gas was inserted.

INTENSITY MEASUREMENTS

A photographic method was used for the measurement of the intensities, the optical set up being that shown in Fig. 3. A glass spectrograph, with an F 2.7 telescope lens was used for the photography. The extreme speed of this instrument made it possible to obtain exposures of satisfactory density with the discharge tube in 10 to 20 minutes, but made the obtaining of a continuous comparison spectrum a matter of some difficulty. In order to obtain a continuous spectrum of sufficiently low intensity, the scheme shown was resorted to. A ribbon filament tungsten lamp was mounted 70 cm from the optical axis. The light from a 3.25 mm section of the middle of the filament (which was 2 mm wide) was allowed to fall on the small right angled prism mounted on the axis. One face of this prism was ground to a focal length of 25 mm, so that a very much reduced image of the filament was formed on the optical axis. This image was used as a secondary source, and only the

⁴ Hodges and Michels, Phys. Rev. **32**, 913 (1928).

light from it intercepted by the slit of the instrument was used for the calibration exposures. The Nicol prism, mounted directly in front of the slit, was arranged so that the two components of polarization could be measured separately and combined later to give the total intensity, thus eliminating errors due to the unequal treatment of the two components by the glass prism.

The 6 to 1 reduction obtained in the spectrograph made it impossible to use the "stepped reducer" method of plate calibration which had been utilized in previous work. It was necessary, therefore, to take a series of five or six calibration exposures. It seemed better, consequently, to eliminate the re-

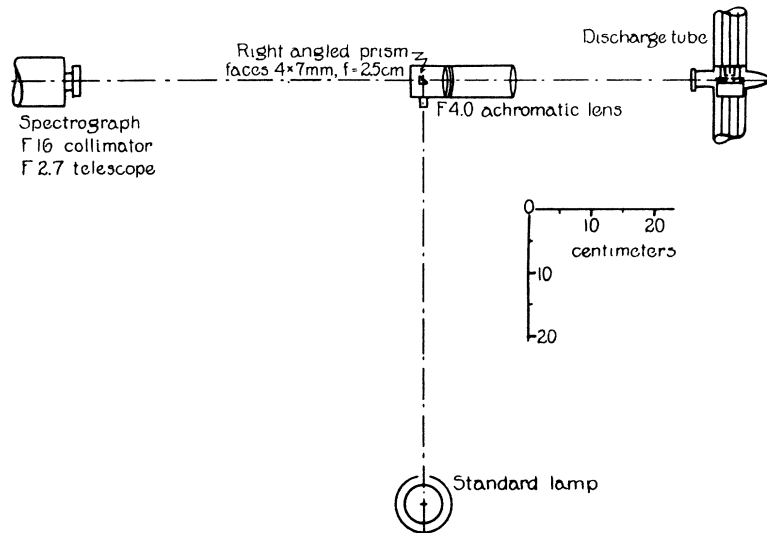


Fig. 3. Optical system.

ducers entirely, and to use a temperature variation of the standard lamp for the intensity steps. This was made particularly convenient by a new method of plotting the calibration curves.

In the older method, the density is plotted against the logarithm of the exposure, or, when the time is held constant, as it was in this work, against the logarithm of the intensity. This plot is widely used because it gives, over a large range of exposures, a straight line, in accordance with the Swartzchild formula. We now recall that tungsten, in the visible region, closely follows Wien's law:

$$I_{\lambda,T} = \epsilon c_1 \lambda^{-5} e^{-c_2/\lambda T}.$$

If, in this formula, we consider the emissivity, ϵ , to be a constant independent of temperature, as it is, very nearly, and vary the temperature, T , for a given wave-length, λ , we find that:

$$\log I_T = a - b/T,$$

where a and b are constants, so that, if the density be plotted against the reciprocal temperature, the straight line of the Swartzchild formula is re-

tained. A typical set of calibration curves for the Ilford panchromatic plates used in this investigation is given in Fig. 4. After the calibration curves had been obtained, the temperature of the tungsten lamp necessary to produce the same density as the helium exposure could be read from them, and the intensity corresponding to this temperature calculated from Wien's law. Correction was then made for the dispersion of the spectrograph to give the final intensities.

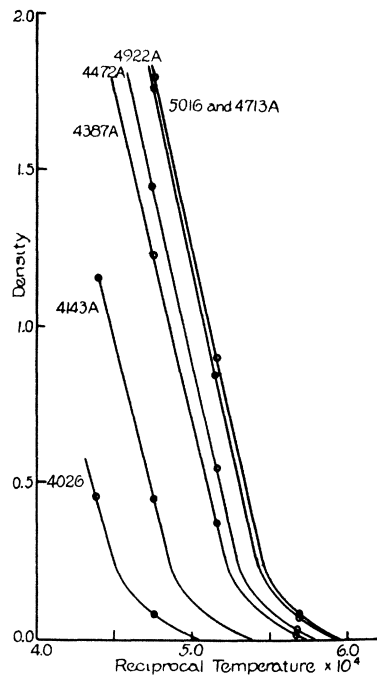


Fig. 4. Plate calibration curves.

The temperatures of the standard lamp were measured with a Leeds and Northrup optical pyrometer, the residual error of which was determined by a Bureau of Standards Calibration to be 2°C . The deviation measure of the temperature measurements was also about 2°C , so the temperature was accurate to about 3°C . Correction for the emissivity of tungsten was made according to the data of Forsythe and Worthing.⁵

It was possible to take fourteen exposures on a single plate. Of these, five, in general, were needed for continuous calibration exposures. The other nine were therefore used for helium spectra, taken under different excitation conditions. The plates were soaked in running water for five minutes, then developed according to the "brush method" recommended by Bloch.⁶ With this treatment, it was found that errors introduced due to difference of posi-

⁵ Forsythe and Worthing, *Astrophys. J.* **61**, 146 (1925).

⁶ Bloch, *Phot. J.* **61**, 425 (1921).

tion on the plate were negligible. The plates were microphotometered in an instrument built at the Institute.

INTENSITY VARIATIONS WITH CURRENT AND PRESSURE

In nearly all previous investigations, tests of the dependence of intensity on the current and pressure have been tried. In most of these tests, a linear relationship has been expected but not found. If we define the excitation function for an isolated atom as was done above, it is obvious that the experimental conditions should be such that the intensity of any line should be proportional to the number of impacts between electrons and the atom in unit time. This number, moreover, is proportional to the current. Also, if the pressure is sufficiently low so that the mean free path is large compared with the linear dimensions of the apparatus, the kinetic theory exponential expression for the number of impacts can be expanded to the first order, and a linear relationship is obtained between the number of impacts and the pressure. From these considerations we should expect, if we have isolated behavior, a linear relationship between either intensity and current or intensity and pressure for any line.

If the condition of isolation is not satisfied, there are several possibilities. In the first place, two atoms, both excited to metastable levels, may collide. In this case, the number of metastable atoms should vary linearly with the current or pressure, so that a quadratic variation of intensity would be expected. Another possibility is emission due to ionization and recombination. For this phenomena we again have the density of ions and the density of electrons each depending linearly on the current and pressure, so we should once more expect a quadratic law. In the general case, then, when some atoms act as isolated, while others are influenced by surrounding atoms, the law of variation between intensity and current or pressure will be a combination of the linear and quadratic variations. In the first work done with the present apparatus, such a law was found to hold, with the quadratic term accounting for about half of the intensity at 100 volts, with a current density of 0.0125 amperes per sq. cm, and a pressure of 0.10 mm of mercury. As the voltage decreased, the importance of the quadratic term also decreased, becoming negligible at about 50 volts.

The difficulty with the above interpretation lies in the fact that the experimentally determined coefficient of the quadratic term is much larger than would be expected. Since the mean distance from any point in the space being observed to the electrodes was of the order of two millimeters, thermal agitation would carry any ion or atom to the wall in $5(10)^{-6}$ seconds. For ions, any positive space charge built up would greatly reduce this time. If the ions are neutralized and the metastable atoms drop back to the ground state, as would be expected on collision with the metal electrodes, the chance for the occurrence of these secondary phenomena would be very small, especially since, with the highest current and pressure ever used, collisions between an electron and any given atom occur once in two or three tenths of a second, and the concentration of metastable atoms, ions and electrons could not attain any very high value.

These difficulties of interpretation were removed by the clean-up treatment outlined above. It was found, after this treatment, not only that a linear relationship was obtained, but also that the constant of proportionality was identical with the coefficient of the linear term previously found. A typical plot of the results finally obtained is shown in Fig. 5, for the line 4472 Å, taken at 100 volts. The spread of the observed points in this figure will also give an idea of the accuracy of measurement possible with the methods used. The variable chosen for the abscissa in this curve is the product of current and pressure. This, obviously, gives the same curve as either, plotted alone, would give, and at the same time saves space.

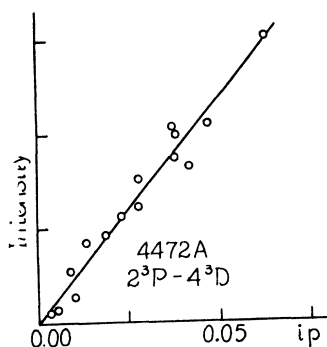


Fig. 5. Intensity variation with current and pressure.

The explanation, possibly, of the change in behavior after cleaning is that the electrodes, when coated with a layer of impurities (stop-cock grease or atmospheric molecules) act very inefficiently for the neutralization of the ions, and allow a large space charge to build up, thus increasing the relative importance of the recombination spectra. Data on the reflection of positive ions from dirty surfaces do not seem to be complete enough to give an answer on this point. Other evidence for the decrease of positive ions after the clean-up was given by the voltage-current curves, which showed no break in passing through the ionization potential.

The linear law finally obtained has been taken as proof that the atoms truly act in an isolated manner, and that contributions due to collisions of the second kind, recombination, etc., have been reduced to negligible proportions.

INTENSITY VARIATIONS WITH VOLTAGE

Figs. 6 and 7 give the intensity-voltage relationships found. These curves were obtained by a series of exposures at pressures between 0.04 and 0.08 mm and with currents of 0.300 to 0.600 ma. By means of the linear relationships established, all these exposures could be reduced to a common pressure and current before plotting. While the relative intensities between the lines are approximately correct, those of wave-length shorter than 4500 Å may be somewhat in error, due to absorption in the glass prism, through which the

line spectra and the continuous light did not have identical paths. This, of course, does not affect the shape of the curve for any individual line, but merely the relative heights.

In Fig. 6 the results for the singlet system are shown, plotted on a logarithmic scale for convenience. The weak lines of the singlet sharp series, while

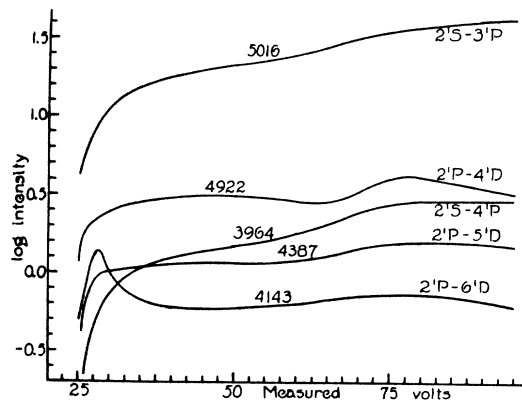


Fig. 6. Intensity-voltage relationships—singlet system.

they were resolved from nearby strong lines, were not far enough removed so that the microphotometer measurements could be relied upon, consequently no results are given for them. The first member of the diffuse series (6678 Å) has also been omitted due to the continuous background from the tungsten filament, which was very appreciable in this region. It will be noticed that

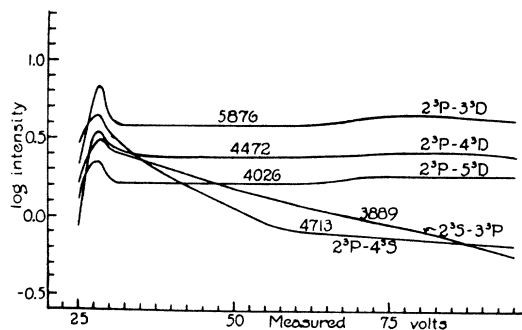


Fig. 7. Intensity-voltage relationships—triplet system.

the two lines observed in the principal series are apparently just approaching maxima at 90 to 100 volts, in quite good agreement with the results of Hanle and Elenbaas. In the diffuse series, 4922 shows two maxima, at 50 and 80 volts, as was found by Elenbaas. The other two lines observed show maxima at 80 volts, and 4143 has a sharp peak at 28 volts. This is the only singlet line showing this low voltage peak. A close examination of the curves will show that, in every case, a point of inflection occurs between 60 and 70 volts, indicating, apparently, some change in the excitation conditions in this region.

The behavior of the triplet system, as shown in Fig. 7, is somewhat different from that of the singlets. At least one member of each series has been observed in this case and all show sharp maxima at 28 volts. The behavior of the diffuse series is remarkably different from the other two, in that the intensity at voltages above the peak does not drop off rapidly. This series also shows maxima at about 80 volts as did the diffuse singlet series. All lines show the same inflection point between 60 and 70 volts as was shown by the singlets.

THE OPTICAL EXCITATION FUNCTION

In previous work, the plot of intensity vs. voltage given above has been taken as the curve of the optical excitation function, on the assumption of an absolutely monochromatic electron beam. With the experimentally determined distribution curve available, however, it is clear that, in the neighborhood of the excitation potential, part of the change in intensity, at least, is due to a change in the number of effective electrons, rather than to a variation in their velocity. The distribution curve shows that 96% of the electrons are included within a range of 10.8 volts. If the excitation potential of a given line, then, is 23 volts, it is clear that, taking into account the 1.2 volt contact potential, the effective current is increasing until a measured voltage of 35 volts has been reached. If the optical excitation function is defined for a monochromatic beam, which is not obtained experimentally, then the intensity will be given by:

$$I = C \int_0^{\infty} F(v) i(V_m, v) dv$$

where C is a constant, $F(v)$ is the optical excitation function for the voltage v , and $i(V_m, v)$ is the current distribution function (i.e. the height of the curve of Fig. 2b) for the same voltage, and for a given value of V_m as defined below. We know, however, that below the excitation potential V_0 the optical excitation function must have the value zero and that above a certain maximum voltage, V_m (1.2 volts below the measured potential), the current distribution function is zero. Making the corresponding change of limits and taking the constant into the function, we have:

$$I = \int_{V_0}^{V_m} F(v) i(V_m, v) dv.$$

Taking the derivative with respect to V_m .

$$\frac{dI}{dV_m} = F(V_m) i_m + \int_{V_0}^{V_m} F(v) \frac{di(V_m, v)}{dV_m} dv$$

where $F(V_m)$ is the value of the excitation function at the maximum voltage and i_m is the height of the flat portion of the distribution curve. Solving for $F(V_m)$ and choosing units so that i_m is unity:

$$F(V_m) = \frac{dI}{dV_m} - \int_{V_0}^{V_m} F(v) \frac{di(V_m, v)}{dV_m} dv.$$

Fortunately, when V_m is less than five volts above the excitation potential di/dV_m is zero throughout the range of integration, and the equation for $F(V_m)$ reduces to the first term. In this range, then, the optical excitation function may be directly determined from the slope of the intensity-voltage curve. This determination supplies a set of values of the function which may be combined with the values of di/dV_m read from Fig. 2a and graphically integrated to give the second term for the computation of the function at voltages not more than ten volts above the excitation potential. The new points thus obtained may be used in turn for calculating still higher voltages.

In Figs. 8 and 9 the results of these computations are shown. While the calculations could not be carried down to the excitation potential itself, due to the large error that would be introduced by a small error in the determina-

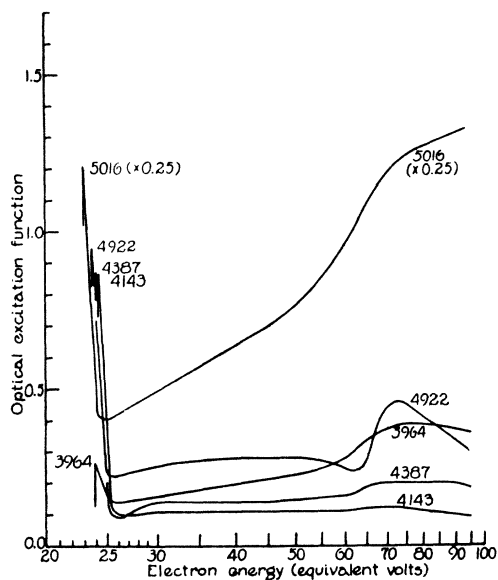


Fig. 8. Optical excitation function—singlet system.

tion of the velocity distribution, they have been carried to within half a volt of that potential. As the optical excitation function is still rising in value at that point, it has been assumed that the peak actually occurs at the excitation potential, and the curves have been extrapolated accordingly. In the figures, a logarithmic scale has been chosen for the voltages and parts of the curves approaching the peaks have been omitted in order to avoid congestion.

It will be noticed that the function, for all lines, exhibits a sharp peak at the excitation potential and a more or less constant value for all higher voltages. The width of the peak itself is, in general, about two volts. Whether

this width is real or is the result of experimental difficulties with voltage control, determination of velocity distribution, etc. cannot be decided with the present data.*

As would be expected from Figs. 6 and 7, the triplets are distinguished from the singlets by the fact that, in general, their ratio of peak height to background height is greater. The singlet principal series seem to be anoma-

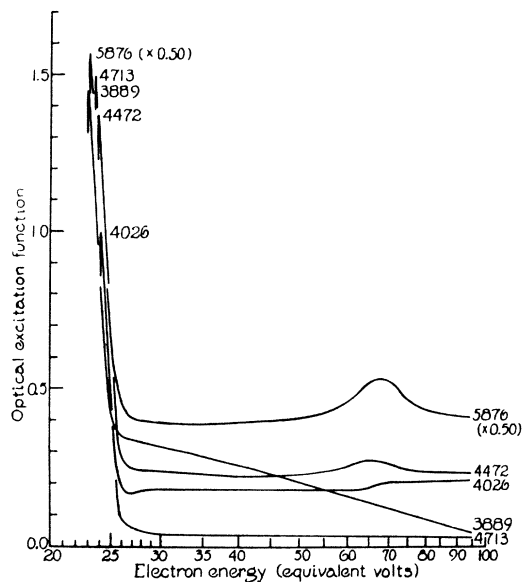


Fig. 9. Optical excitation function—triplet system.

lous in that its background value is of the same order of magnitude as the height of its maxima, and the two members observed show a much greater ratio of intensities than do the succeeding members of any other series. This may be connected with the fact that the normal state of the atom is a 1S level, with allowed transitions to the 1P levels.

CONCLUSIONS

We may summarize the new facts in regard to the optical excitation function of helium which have been brought forward in this investigation:

(1) It is possible, by proper apparatus and technique, to obtain experimental conditions under which the atoms act in an isolated manner, with no appreciable effect of recombination. The test for this condition is a linear dependence of intensity on either current or pressure, within a limited range.

(2) The optical excitation function has been shown to have a similar behavior for all lines, in that a sharp maxima is observed either at or very near to the excitation potential.

* It would seem probable that this peak may actually consist of a series of maxima, here unresolved, corresponding to the various excitation levels of the helium atom.

(3) Between 60 and 70 volts an irregularity occurs in the function, possibly due to some new mode of excitation entering in this region.

(4) The singlet and triplet systems are distinguished by the fact that, in the former, excitation is accomplished with fairly high probability by electrons with all energies above that just necessary for excitation, while, in the latter, those electrons with just sufficient energy to excite are much more efficient than those with greater energy.

In conclusion the author wishes to express his extreme gratitude to Drs. R. A. Millikan, I. S. Bowen, and W. V. Houston, whose constant interest has been of great aid in the completion of this investigation. He also wishes to thank Mr. A. B. Anderson for assistance in taking some of the data.