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ON THE MEASUREMENTS OF LIFETIMES, EXCITATION FUNCTIONS, DENSITIES, TOTAL CROSS SECTIONS, AND POLARIZATION RATIOS OF THE METASTABLE STATE OF THE NEON ATOM UNDER ELECTRONIC IMPACT

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Author

Hadeishi, Tetsuo.

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Ernest O. Lawrence
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Tetsuo Hadeishi

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ABSTRACT

The values of lifetimes, excitation functions, densities, total cross sections, and polarization ratios of the metastable state ($2^2P_{3/2}$ $3^2S_{1/2}$)₂ of neon at various pressures under electronic impact have been measured by use of an electron gun. A method of obtaining the total cross section from the measurement of lifetime, electron flux, excitation function, and width of the resonance line is derived and discussed.

We found two apparent lifetimes of the metastable state, one of which is much shorter than the other at the same potential. The longer one is in agreement with previously published results; however, the shorter one is not. We interpret the longer lifetime to be caused by high lying metastable states feeding into the lower metastable state.

We found that the shorter lifetime is less sensitive to the variation of pressure. The observed half-lives are 0.428(17), 0.452(33), 0.497(16), and 0.451(36) msec at the pressure settings of 0.41, 0.69, 1.03, and 1.68-mm Hg, respectively, at 300°K (the numbers in parentheses indicate a plus or minus error in the last two digits). The measured values of the half-lives corresponding to the longer lifetime are 2.5(1.8), 4.8(4.0), 8.3(7.5) msec for the pressure settings of 1.68, 1.03, and 0.69-mm Hg at 300°K. The measured average value of the density of the metastable-state atoms is $5.3(1.5) \times 10^9/\text{cm}^3$. The

measured average total cross section is $1.2(0.6) \times 10^{-19} \text{ cm}^2$. The polarization ratio--defined by $(\Delta I_{\perp} - \Delta I_{\parallel})/(\Delta I_{\perp} + \Delta I_{\parallel})$, where $\Delta I_{\parallel}$ and $\Delta I_{\perp}$ are the intensities of the absorbed light when the electric vector is parallel to and perpendicular to the direction of the electron beam--showed a positive value only at the excitation potential very close to the threshold of excitation. The polarization ratios measured were negative for excitation potentials higher than 2V above the threshold of excitation. Even though we obtained a negative polarization ratio for electron energies greater than 19V, we believe this can not be used for rf resonance since we cannot expect to increase this polarization ratio any more than what we have observed under this energy range.

We present details of the apparatus used to measure these quantities associated with the metastable state of the neon atom, and discuss our results.

I. INTRODUCTION

Several studies have been made concerning the lifetimes of metastable states of noble gases. The general features of the metastable atoms of helium, neon, and argon were established through these experiments.¹⁻⁴ All these previous experiments involved detecting the metastable-state density in the afterglow of an interrupted discharge by means of optical absorption of appropriate wave lengths.

We have produced the metastable states of neon by electronic impact on ground state atoms, and have detected the metastable states by means of the optical absorption line at $6143\text{-}\text{\AA}$. The relevant energy levels of a neon atom used in the experiment are shown in Fig. 1. The excitation electrons were produced by the electron gun and because of this we were able to turn the electron beam "on" and "off" and vary the energy of the electrons. We shine the $6143\text{-}\text{\AA}$ monitoring light perpendicular to the direction of the electron beam. The amount of absorption of this light is proportional to the density of the metastable-state atoms produced by the electron beam. Therefore, when the production of the metastable states is terminated by applying a negative potential to the grid of the electron gun, the absorption of the $6143\text{-}\text{\AA}$ light will change as a function of time. If the intensity of the $6143\text{-}\text{\AA}$ light is small, the depopulation of the metastable-state atoms by the $6143\text{-}\text{\AA}$ light will be small. By noting the decrease in the absorption of the $6143\text{-}\text{\AA}$ line, we get information concerning how fast the metastable-state atoms are decaying. This is the basis for the lifetime measurement.

One of the disadvantages of previous experiments employing the interrupted discharge is the appearance of a Faraday dark space which prevents lifetime measurements immediately after the termination of discharge. This effect was investigated by Phelps and Molnar⁴ who interpreted it as being caused by the presence of electrons after the spark was terminated. These electrons destroy the metastable states through excitation or ionization.

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Fig. 1. Relevant energy levels of a neon atom used in the experiment. The first excited level is the metastable state with $J=2$ produced by electronic impact. The 6143-Å line connecting the two excited levels is the one used to monitor the behavior of the metastable state.

In our experiment we shut off the electron beam very rapidly by a negative potential to the grid of the electron gun; we are then able to measure the relaxation effect of the metastable states immediately following the termination of the electron beam. This reveals a considerable discrepancy from the measured lifetimes of previous experimental results. We interpret the longer lifetime observed in the previous experiments as being due to the feeding of the metastable states by higher metastable states. Each absorption curve corresponding to a particular pressure setting reveals two lifetimes. The one with a shorter lifetime shows a rather small variation with respect to a change in pressure, whereas the one with the longer lifetime shows a marked change with a change in pressure; the long lifetime increases as the pressure becomes lower. This effect indicates the existence of the long-lived higher metastable states that, as a result of collision of atoms, relax to the metastable state of our interest. It is the longer lifetime which agrees with the previously published results. Indeed, this is the time region in which Phelps and Molnár⁴ made their measurement.

We measure, in addition to the lifetime, the excitation function, the absolute density of the metastable states under the electronic impact, the total excitation cross section, and the polarization ratio. We determine the excitation function by measuring the amount of light absorption as a function of electron energy. The observed quantity is in good agreement with that published by Milatz and Orenstein.⁵

Knowing the excitation function and the width of the resonance radiation of the 6143-Å line, we determine the absolute density of the metastable states as a function of the excitation energy of the electrons. Details of theory behind our method are presented in Sec. II. We derive a method of obtaining the total cross section using additional measurements of electron-current density. The total cross section is obtained using this method. The details of the derivation are also given in Sec. II.

The polarization ratio is defined by $P = (\Delta I_{\perp} - \Delta I_{\parallel}) / (\Delta I_{\perp} + \Delta I_{\parallel})$ where $\Delta I_{\parallel}$ and $\Delta I_{\perp}$ are the absorbed intensities of the linearly polarized light transmitted through the electron-gun interaction region when the direction of the polarization is perpendicular to and parallel to the direction of the electron beam, respectively. To observe a radio-frequency resonance of the metastable states it is necessary for this ratio to be as large as possible.

Originally, we undertook this research to investigate the possibility of observing an rf resonance of the metastable states produced by electronic impact; we hoped that a sufficient amount of the metastable state $(2^2P_{3/2} \ 3^2S_{1/2})_2$ of neon would be aligned. Unfortunately, we found the alignment effect in our experiment to be too small for us to see the rf - resonance effect. We think that a much lower density of neon is required for increasing the alignment. * However, when we lower the pressure of the neon, the absorption goes down below the detection capability of our detector. One suggestion is to make a cathode over 30 cm long and to shine the light parallel to it in order to increase the absorption path length. * Such a setup is now under consideration.

We believe that considerable information about the metastable state of neon has been obtained from our experiment, which may be of some help in observing an rf resonance of noble-gas atoms in the near future.

*Private communication with Professor W. R. Bennett, Physics Department, Yale University.

II. THEORY

If parallel light from a source that emits a continuous spectrum is sent through an absorption cell containing a monatomic gas, the intensity of transmitted light I_ν may show a frequency distribution similar to that in Fig. 2. The absorption coefficient k_ν of the gas is defined by the equation

$$I = I_0 \exp(-k_\nu x), \quad (1)$$

where x is the thickness of the absorbing layer. From Eq. (1) we may obtain k_ν as a function of frequency as shown in Fig. 3.

Consider a parallel beam of light of frequency between ν and $\nu + d\nu$ and I_ν traveling in the x direction, through a layer of atoms bounded by the plane x and $x + dx$. Suppose there are N atoms/cm³ of which δN_ν are capable of absorbing the frequency between ν and $\nu + d\nu$, and N' excited atoms of which $\delta N'$ are capable of emitting this frequency range. Let us first consider two energy levels denoted by E_i and E_j , such that $E_j > E_i$. If we neglect the effect of spontaneous re-emission because the emitted radiation is isotropically distributed and because the possibility of spontaneous re-emission may take place to other levels than that from which the initial absorption has taken place, the decrease in beam energy is given by

$$-d[I_\nu \delta \nu] = \delta N_\nu dx h \nu B(i, j) \frac{I_\nu}{4\pi} - \delta N' dx h \nu B(j, i) \frac{I_\nu}{4\pi}, \quad (2)$$

where $B(i, j)$ is the Einstein stimulated transition probability from the level i to j . Originally, the Einstein B coefficient was defined in terms of radiation density. However, for experimental purposes it is convenient to express it in terms of isotropic radiation intensity.

The relation between the B coefficient defined in terms of radiation density and the isotropic radiation intensity is $B(\text{density}) = \frac{c}{4\pi} B(\text{intensity})$.³ From Eq. (2) we find

$$-\frac{1}{I_\nu} \frac{dI_\nu}{dx} \delta \nu = \frac{h\nu}{4\pi} [B(i, j)\delta N - B(j, i)\delta N']. \quad (3)$$

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Fig. 2. An absorption line.

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Fig. 3. Variation of absorption coefficient with frequency in an absorption line.

From Eq. (1), we recognize that

$$k_\nu d\nu = \frac{h\nu}{4\pi} [B(i, j)\delta N - B(j, i)\delta N'] .$$

If we assume that variation in ν in $h\nu$ is small over the region of integration, then we have

$$\int k_\nu d\nu = \frac{h\nu_{ij}}{4\pi} [B(i, j)N - B(j, i)N'] , \quad (4)$$

where

$$h\nu_{ij} = |E_j - E_i| .$$

With the assumption that $N \gg N'$, we get

$$\int k_\nu d\nu = \frac{h\nu_{ij}}{4\pi} B(i, j) N . \quad (5)$$

Let $\Delta I/I$ be the fractional reduction of intensity of the radiation over the absorption line. Then we have

$$\frac{\Delta I}{I} = 1 - \frac{\text{Transmitted intensity}}{\text{Incident intensity}} .$$

Using Eq. (1), we obtain

$$\frac{\Delta I}{I} = \frac{\int_{-\infty}^{\infty} I_0(\nu) (1 - e^{-k_\nu \ell}) d\nu}{\int_{-\infty}^{\infty} I_0(\nu) d\nu} , \quad (6)$$

where ℓ is the optical path length through which the incident radiation passes. If we make the assumption that the absorption is small

$$\frac{\Delta I}{I} = \frac{\int_{-\infty}^{\infty} I_0(\nu) k_\nu d\nu}{\int_{-\infty}^{\infty} I_0(\nu) d\nu} . \quad (7)$$

Since the measured absorption is only about 2%, the above approximation is justified. Spectral distribution $I_0(\nu)$ corresponds to the spectral distribution of the resonance radiation from the lamp. The measured value of the half-width for the neon line 6143-Å which corresponds to the transition between the states $(2^2P_{3/2} 3^2S_{1/2})_2$ to $(2^2P_{3/2} 3^2P_{1/2})_2$ is 0.0364-Å which corresponds to 2880 Mc/sec. The half-width of the coefficient of absorption is about 110 Mc/sec. This is about 1 part in 26. So if we plot $I_0(\nu)$ and k_ν vs frequency we get curves like the ones shown in Fig. 4.

Since the variation of $I_0(\nu)$ over the frequency ν_0 is very small compared with the variation of k_ν we may approximate Eq. (7) by

$$\frac{\Delta I}{I} = \frac{\ell \int_{-\infty}^{\infty} k_\nu d\nu}{\Delta \nu}, \quad (8)$$

where $\Delta \nu$ is the resonance width of the lamp. The validity of this approximation is discussed in Appendix I. From Kramer-Heisenberg's dispersion formula,⁶ we get

$$B(\text{density}) = \frac{\pi e^2}{m_0 h \nu_{ij}} \left| f_{ij} \right|$$

where f_{ij} is the oscillator strength for the transition from the state i to j . From the relation

$$B(\text{intensity}) = \frac{4\pi}{C} B(\text{density})^3$$

we obtain

$$B(\text{intensity}) = \left(\frac{4\pi}{C} \right) \frac{\pi e^2}{m_0 h \nu_{ij}} \left| f_{ij} \right| \quad (9)$$

Substituting it into Eq. (5), we get

$$\int_{-\infty}^{\infty} k_\nu d\nu = \frac{\pi e^2 N}{m_0 C} \left| f_{ij} \right|. \quad (10)$$

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Fig. 4. Comparison of relative width of the line from the lamp with the coefficient of absorption.

Therefore, Eq. (8) becomes

$$\frac{\Delta I}{I} = \left(\frac{l}{\Delta \nu} \right) \frac{\pi e^2 N}{m_0 C} \left| f_{ij} \right|. \quad (11)$$

Therefore, from the measurement of $\frac{\Delta I}{I}$, $\Delta \nu$, and the oscillator strength f_{ij} , we are able to determine the density of the metastable state N .⁷

The rate equation for the metastable state whose density is N is expressed by

$$\frac{dN}{dt} = \sigma_t \left(\frac{J}{e} \right) N_0 - \frac{1}{\tau} N, \quad (12)$$

where

J/e (the ratio of current density to electronic charge) corresponds to the flux of the electron beam, τ is the lifetime of the metastable state, and σ_t is the total cross section. The flux of the electron beam may be shut off electrically by applying a negative voltage to the grid of the gun. When the electron beam is shut off at $t = t_0$, the density of the metastable state as a function of time is given by

$$N(t) = N(t_0) \exp(-t/\tau). \quad (13)$$

The decay constant τ is measured from the slope of the absorption curve displayed on the cathode-ray oscilloscope. At equilibrium, $dN/dt = 0$. Hence, we obtain

$$\sigma_t = \frac{(N/\tau)}{(J/e)N_0}. \quad (14)$$

From Eq. (11), we get

$$N = \frac{1}{(l/\Delta \nu) \frac{\pi e^2}{m_0 C} \left| f_{ij} \right|} \times \left(\frac{\Delta I}{I} \right). \quad (15)$$

Combining Eqs. (14) and (15), we get

$$\sigma_t = \frac{1}{\left(\frac{J}{e}\right)N_0} \times \frac{1}{\left(\frac{l}{\Delta\nu}\right)\frac{\pi e^2}{m_0 C} |f_{ij}|} \times \left(\frac{\Delta I}{I}\right) \times \frac{1}{\tau} . \quad (16)$$

Surprisingly accurate values consistent with the experiment are obtained by the method of Bates and Damgaard⁸ for the oscillator strength of Li I, O II, Si II, Ca II, and Hg I when one uses the coulomb approximation.

$$\left(\int_0^\infty R_i R_j r dr \right)^2$$

where R_i/r and R_j/r are, respectively, the initial and final radial wave function of the active electron normalized in atomic units in the Bates and Damgaard method. The details of their method are given in Appendix II. The oscillator strength is defined by

$$f_{ij} = \frac{8\pi^2 m_0}{3 e^2 h} \left| \langle i | \underline{p} | j \rangle \right|^2 , \quad (17)$$

where

$$\left| \langle i | \underline{p} | j \rangle \right|^2 = \left[\left| \langle i | \sum_k e x_k | j \rangle \right|^2 + \left| \langle i | \sum_k e y_k | j \rangle \right|^2 + \left| \langle i | \sum_k e z_k | j \rangle \right|^2 \right] .$$

Now, we have the quantities

$$C_q^k = (-1)^q \sqrt{\frac{(k-q)!}{(k+q)!}} p_k^q (\cos \theta) e^{iq\phi} , \quad (18)$$

with

$$P_q^k = \sin^q \theta \frac{d^q}{d(\cos \theta)^q} P_k(\cos \theta) ,$$

where

$$P_k(x) = \frac{1}{2^k k!} \frac{d^k}{dx^k} (x^2 - 1)^k$$

satisfies the conditions of irreducible tensor operator of rank k defined by Racah.⁹ We shall make use of his method for calculating the oscillator strength. It can be shown from Eq. (18) that

$$C_{\pm 1}^1 = \mp \sqrt{\frac{1}{2}} \frac{(x \pm iy)}{r} \quad \text{and} \quad C_0^1 = \frac{z}{r}.$$

Therefore, we have

$$x = \frac{r}{\sqrt{2}} [C_{-1}^1 - C_{+1}^1], \quad y = \frac{r}{i\sqrt{2}} [C_{-1}^1 + C_{+1}^1],$$

and

$$z = r C_0^1.$$

Hence, we may write Eq. (18) as

$$\begin{aligned} |\langle i | \underline{p} | j \rangle|^2 = & \frac{e^2}{2} \left[\left| \langle i | \sum_k r_k (C_{-1}^1 - C_{+1}^1)_k | j \rangle \right|^2 \right. \\ & + \left| \langle i | \sum_k r_k (C_{-1}^1 + C_{+1}^1)_k | j \rangle \right|^2 \\ & \left. + \left| \langle i | \sum_k r_k (C_0^1)_k | j \rangle \right|^2 \right] \end{aligned} \quad (19)$$

where

$$\langle i | = \sum_{M_J} \langle \gamma(2^2 P_{3/2}^5, 3^2 S_{1/2}^1) J, M_J | = \sum_{M_J} \langle \gamma(2^2 P_{3/2}^5, 3^2 S_{1/2}^1) 2, M_J |$$

corresponding to the metastable state and

$$|j\rangle = \sum_{M_J'} |\gamma(2^2 P_{3/2}^5, 3^2 P_{1/2}^1) 2, M_J', \rangle$$

corresponding to the excited state with the optical transition with 6143-Å. Hence, we obtain

$$\begin{aligned} |\langle i | \underline{p} | j \rangle|^2 &= e^2 \left(\int_0^\infty R_1 R_2 r dr \right)^2 \\ &\times \sum_{M_J} \sum_{M_{J'}} \left[\left| \langle (j_1 j_2) J, M_J | C_{-1}^1 | (j_1 j_2') J, M_{J'} \rangle \right|^2 \right. \\ &+ \left| \langle (j_1 j_2) J, M_J | C_{+1}^1 | (j_1 j_2') J, M_{J'} \rangle \right|^2 \\ &+ \left. \left| \langle (j_1 j_2) J, M_J | C_0^1 | (j_1 j_2') J, M_{J'} \rangle \right|^2 \right] \end{aligned} \quad (20)$$

where j_1 and j_2 and j_2' correspond to the parent term $^2P_{3/2}$, and the active electron term $^2S_{1/2}$ and $^2P_{1/2}$. Hence, the matrix elements to be evaluated are of the form

$$\langle j_1 j_2 J M_J | X_Q^K | j_1 j_2' J M_{J'} \rangle \quad (21)$$

Using the Wigner-Eckart theorem,¹⁰ we have

$$\langle j_1 j_2 J M_J | X_Q^K | j_1 j_2' J M_{J'} \rangle = (-1)^{J-M_J} \begin{pmatrix} J & K & J' \\ -M_J & Q & M_{J'} \end{pmatrix} \langle j_1 j_2 J || X^K || j_1 j_2' J \rangle$$

where $\begin{pmatrix} J & K & J' \\ -M_J & Q & M_{J'} \end{pmatrix}$ is the 3-j symbols defined in Edmonds¹¹ and

$\langle j_1 j_2 J || X^K || j_1 j_2' J \rangle$ is the reduced matrix element. It can be shown that when the dipole operator

$$\underline{p} = e \sum_{\underline{k}} \underline{r}_{\underline{k}}$$

is separated into two parts--one of which operates on the parent term and the other on the terms corresponding to $^3S_{1/2}$ and $^3P_{1/2}$ --the only non-vanishing part is the one operating on the latter terms corresponding to the active electron terms.

Now, $X_Q^K \equiv \left\{ \begin{matrix} k_1 & k_2 \\ \underline{U} & \underline{T} \end{matrix} \right\}_Q^K$ with $\underline{U}^{k_2} = 1$. Hence

$$X_Q^K = T_{q_2}^{k_2} = T_q^1. \text{ Therefore, } \langle j_1 j_2^J \| X_Q^K \| j_1 j_2'^J \rangle = \langle j_1 j_2'^J \| T_q^1 \| j_1 j_2^J \rangle$$

with T^1 operating only on the part 2. Therefore, we have

$$\langle \gamma j_1 j_2^J \| T^1 \| \gamma j_1 j_2^J \rangle = (-1)^{j_1+j_2+J+1} \sqrt{(2J+1)(2J+1)} \\ \times \left\{ \begin{matrix} J & 1 & J \\ j_2' & j_1 & j_2 \end{matrix} \right\} \langle \gamma j_2 \| T^1 \| \gamma j_2 \rangle$$

where the 6-j symbol $\left\{ \begin{matrix} J & 1 & J \\ j_2' & j_1 & j_2 \end{matrix} \right\}$ is also defined in Edmonds.¹¹ We

evaluate $|\langle i | p | j \rangle|^2$ using the above formula. The matrix elements to be evaluated are

$$\sum_{M_J} \sum_{M_J'} \sum_q \left| \langle (j_1 j_2)^{JM_J} | C_q^1 | (j_1 j_2')^{JM_J'} \rangle \right|^2 \\ = \sum_{M_J} \sum_{M_J'} \sum_q \left(\begin{matrix} J & 1 & J \\ -M_J & q & M_J' \end{matrix} \right)^2 \left| \langle j_1 j_2^J \| C_q^1 \| j_1 j_2'^J \rangle \right|^2 \\ = \sum_{M_J} \sum_{M_J'} \sum_q \left(\begin{matrix} J & 1 & J \\ -M_J & q & M_J' \end{matrix} \right)^2 (2J+1) \left\{ \begin{matrix} J & 1 & J \\ j_2' & j_1 & j_2 \end{matrix} \right\}^2 \left| \langle \gamma j_2 \| C^1 \| \gamma j_2' \rangle \right|^2$$

The reduced matrix elements $\langle \gamma j_2 \| C^1 \| \gamma j_2' \rangle$ still require further reduction since C^1 operates only on the orbital part of the wave function.

Now $\langle j_2 | = \langle (s, \ell) j_2 |$ and $| j_2' \rangle = (s', \ell') j_2' \rangle$. Therefore,

$$\langle (s, \ell) j_2 \| C^1 \| (s', \ell') j_2' \rangle \\ = \delta(s, s') (-1)^{s'+\ell'+j_2'+1} \sqrt{(2j_2+1)(2j_2'+1)} \left\{ \begin{matrix} j_2 & 1 & j_2' \\ \ell' & s & \ell \end{matrix} \right\} \langle \ell \| C^1 \| \ell' \rangle$$

with

$$\langle \ell \| C^1 \| \ell' \rangle = (-1)^\ell \sqrt{(2\ell+1)(2\ell'+1)} \begin{pmatrix} \ell & 1 & \ell' \\ 0 & 0 & 0 \end{pmatrix}$$

Therefore, we finally get

$$\begin{aligned} & \sum_{M_J} \sum_{M_{J'}} \sum_q |\langle (j_1 j_2) J, M_J | C_q^1 | (j_1 j_2') J, M_{J'} \rangle|^2 \\ &= \sum_{M_J} \sum_{M_{J'}} \sum_q \left(\begin{matrix} J & 1 & J \\ -M_J & q & M_{J'} \end{matrix} \right)^2 (2J+1)^2 \left\{ \begin{matrix} J & 1 & J \\ j_2' & j_1 & j_2 \end{matrix} \right\}^2 (2j_2+1)(2j_2'+1) \\ & \quad \times \left\{ \begin{matrix} j_2 & 1 & j_2' \\ \ell' & s & \ell \end{matrix} \right\}^2 \delta(s, s') (2\ell+1)(2\ell'+1) \begin{pmatrix} \ell & 1 & \ell' \\ 0 & 0 & 0 \end{pmatrix}^2. \end{aligned}$$

Now $R_i = R_{3,0}$, $R_j = R_{3,1}$, $J = 2$, $j_1 = 3/2$, $j_2 = 1/2$, $j_2' = 1/2$, $s = 1/2$, $\ell = 0$ and $\ell' = 1$. Therefore, we have

$$\begin{aligned} |\langle i | \underline{p} | j \rangle|^2 &= e^2 \left(\int_0^\infty R_{3,0} R_{3,1} r dr \right)^2 \sum_{M_J} \sum_{M_{J'}} \sum_q \left(\begin{matrix} 2 & 1 & 2 \\ -M_J & q & M_{J'} \end{matrix} \right)^2 (25)(12) \\ & \quad \times \left\{ \begin{matrix} 2 & 1 & 2 \\ \frac{1}{2} & \frac{3}{2} & \frac{1}{2} \end{matrix} \right\}^2 \times \left\{ \begin{matrix} \frac{1}{2} & 1 & \frac{1}{2} \\ 1 & \frac{1}{2} & 0 \end{matrix} \right\}^2 \times \begin{pmatrix} 0 & 1 & 1 \\ 0 & 0 & 0 \end{pmatrix}^2. \end{aligned}$$

$$\text{Now, we have } \left\{ \begin{matrix} 2 & 1 & 2 \\ \frac{1}{2} & \frac{3}{2} & \frac{1}{2} \end{matrix} \right\}^2 = \frac{1}{20}, \begin{pmatrix} 0 & 1 & 1 \\ 0 & 0 & 0 \end{pmatrix}^2 = \frac{1}{3}$$

$$\text{and } \left\{ \begin{matrix} \frac{1}{2} & 1 & \frac{1}{2} \\ 1 & \frac{1}{2} & 0 \end{matrix} \right\}^2 = \frac{1}{6} \text{ from the formula from Edmonds.}^{11}$$

Therefore, we get

$$\begin{aligned} |\langle i | \underline{p} | j \rangle|^2 &= e^2 \left(\int_0^\infty R_{3,0} R_{3,1} r dr \right)^2 \sum_{M_J} \sum_{M_{J'}} \sum_q \frac{5}{6} \left(\begin{matrix} 2 & 1 & 2 \\ -M_J & q & M_{J'} \end{matrix} \right)^2 \\ &= \frac{5}{9} e^2 \left(\int_0^\infty R_{3,0} R_{3,1} r dr \right)^2. \quad (21) \end{aligned}$$

Hence, combining with Eq. (17) we get

$$f_{ij} = \frac{8 \pi^2 m_0}{3 e^2 h} \times \frac{5}{9} e^2 \left(\int_0^{\infty} R_i R_j r dr \right)^2 . \quad (22)$$

The calculated value for the oscillator strength $f_{ij} = 0.22$. This is in fairly close agreement with the experimental value obtained by Landenburg and Levy¹² of 0.18. Because of the good accuracy that the Bates and Damgaard method yields for many other atoms, we shall use the calculated value rather than the experimental value observed in 1931 with somewhat crude apparatus.

The polarization ratio was obtained by measuring the difference of absorption when the electric vector of the incident resonance radiation is parallel to and perpendicular to the direction of electron beams as shown in Fig. 5.

The polarization ratio is defined by

$$P = \frac{\Delta I_{\perp} - \Delta I_{\parallel}}{\Delta I_{\perp} + \Delta I_{\parallel}}$$

where $\Delta I_{\parallel}$ and $\Delta I_{\perp}$ are the intensities of the absorbed light when the electric vector of the incident light is parallel and perpendicular to the direction of the electron beams. The result we obtained from the experiment shows that the polarization ratio is too small for us to expect to see the rf resonance by means of our present apparatus.

MU-28332

Fig. 5. Direction of lights and electron beam.

III. EXPERIMENTAL ARRANGEMENT

Standard electronics are used throughout the experiment. An overall schematic of the essential parts of the experiment is shown in Fig. 6.

A. Electron Gun

A schematic of the electron gun is shown in Figs. 7 and 8. It is a tetrode in construction. After three failures, we decided to simplify the design as much as possible and to make the gun as heavy duty as possible. We decided to use a Phillip's cathode, which is barium oxide imbedded in molybdenum. This cathode can withstand the bombardment of positive ions produced by the energetic electron impact. The oxide-coated cathodes used in previous guns lasted only a few minutes due to the positive-ion impact. Since the size of the Phillip's cathode is so large, it has a large heat capacity. None of the heater wires commercially available were able to heat up the cathode to the activation temperature. First we tried to heat it by electron impact. However, we realized that even though this is the simplest way to heat the cathode to the very high temperature required, the whole gun would light up when the neon gas is introduced. Therefore, we had no other choice but to make a wire which would heat the cathode indirectly. We tried various schemes such as noninductively winding a tungsten wire on the ceramic spool, etc., but the wire always burned out before the cathode reached activation temperature. Finally, we wound 15-mil tungsten wire noninductively and coated it with Alumina, which has a very high melting point. By using this heater wire, and by thoroughly shielding the cathode to reduce radiated heat loss, we could finally heat the cathode to the activation temperature and operate it for a few hundred hours.

The rest of the operation for constructing the gun was relatively simple and straight forward. Details of the gun are given in Figs. 7 and 8. However, it was necessary to take the gun apart quite frequently.

MU-28333

Fig. 6. Schematic diagram of experimental system.

MU-28334

Fig. 7. Electron-gun base.

MU-28335

Fig. 8. Cross sectional view of the electron gun.

For this reason, we constructed the gun so that the glass envelope (see Fig. 7) can be removed simply by taking out 12 bolts holding its flange.

We made the electron gun to be bakable. The electron beam was chopped by applying a negative potential to the extraction grid of the electron gun. The response of the electron beam with respect to the application of the negative voltage was observed by monitoring the shape of the collector current by means of a cathode-ray oscilloscope and it is found that the response is less than a μ sec. Since we are measuring lifetimes in the range of msec, this response time is sufficient for our purpose. Electron beam flux is obtained by a measurement of the collector current. We noticed that the grid voltage varied as we increase the electron energy. Therefore, the grid voltage was continuously monitored by the oscilloscope and kept at a constant value to ensure that the characteristic curve of the electron gun was faithfully followed.

B. Neon Lamp Optical System and Detection

The neon lamp is excited by means of an rf field at 80 Mc/sec. To get as much light as possible from the lamp, we constructed the rf oscillator by using two Eimac 4X 250B power tubes which have a maximum rating of 450W each operating as a multivibrator. The details of the circuit diagram are shown in Fig. 9.

Details of the component used in the oscillator are given in Table I. Various types of lamp bulbs have been constructed and tested in addition to the commercially available bulbs. We found that the commercially produced Geisler-type tube is just as good if not better than those constructed by this laboratory. For this reason, most of our experiments were run using the commercially produced bulbs filled with neon.

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Fig. 9. Circuit diagram of the lamp oscillator.

Table I. Components used in the rf oscillator

Item	Value
C_1 and C_2	$C_1 = C_2$ Air gap of wire placed near the plate of the tube. The gap is adjusted until the oscillation of the oscillator is established.
C_4	1000 μmf
C_5 and C_6	Variable capacitor. To be adjusted until the desired operating condition for the oscillation is established. 1500 μmf max.
L_1	8 turns, 1-1/2-in. diam and 3-1/2-in. long. Dimension not critical.
L_2	5 turns, 1-in. diameter and 2-in. long. Dimension not critical.
L_4 and L_5	$L_4 = L_5$. Used as a choke but not essential.
R_1 and R_2	100 Ω . Not essential.
R_3 and R_4	2500 Ω , 15W. Essential for the protection of the screen.
R_5	2500 Ω , 15W. Desirable for obtaining the desired grid bias.

We use an interference filter on the light from the lamp. The interference filter has a half-width of $15 \text{ \AA}$ and is produced by Spectrolab, Inc. This filter is used to select the $6143\text{-}\text{\AA}$ line before the light enters the interaction region. The interaction region is the gap between the screen and collector plate in the electron-gun structure. When we analyze the light through the interference filter by means of monochromator and high-resolution grating we find that in addition to the $6143\text{-}\text{\AA}$ line, there are three more lines corresponding to $6164\text{-}\text{\AA}$, $6128\text{-}\text{\AA}$, and $6096\text{-}\text{\AA}$. Because of this, it is necessary to place a high-resolution monochromator after the electron gun to select only the $6143\text{-}\text{\AA}$ line. However, when the monochromator is used, the light suffers a large attenuation. This is a reason for constructing the high-power oscillator. Also, because of the radiation caused by the spontaneous emission of neon originating from the electron gun itself, we place two converging lenses in front of the monochromator so that the lens system focusses the light from the lamp on the entrance slit of the monochromator; then the light originating in the gun suffers a large dispersion caused by the two lenses before it enters the monochromator. A linear polarizer of polaroid sheet HM-38 whose transmittance $k_1 = 0.79$ and $k_2 = 0.003$ at $6000 \text{ \AA}$ is used. For all practical purposes we may assume it to be a perfect polarizer.

To increase the signal-to-noise ratio, a phase-sensitive detector is used for excitation function measurements. The "lock-in" is easily accomplished by driving the square-wave generator used for chopping the electron beam in synchronism with the internal oscillator of the phase-sensitive detector itself. For measurements of lifetimes, current from the photomultiplier tube is displayed directly on the cathode-ray oscilloscope. The traces of lifetimes were photographed by a scope camera and were enlarged for ease of reading. The points taken from the photograph were programmed and processed by an IBM 7090 computer for the lifetime measurements. A schematic diagram showing the neon lamp, optical system, and detection system is shown in Fig. 10. Table II lists the essential equipment used in Fig. 10.

Table II. Table of essential equipment used for neon lamp, optical, and detection systems

Item	Manufacturer	Model	Function
<u>Power supply</u>			
A	Lamda Electronic Corp. , Corona, N. Y.	32	Power supply A, B, and C in series for necessary plate voltage to operate rf oscillator (1200 V)
B	Lamda Electronic Corp. , Corona, N. Y.		
C	Electronic measurements Co. Redbank, N. J.		
<u>Power supply</u>			
E	NJE Corp. , Kenilworth, N. J.	CS 64 H-40	Provides a negative high voltage for photomultiplier tube.
<u>Power supply</u>			
F	Lamda Electronic Corp. , Corona, N. Y.	28	Provides 250 V for screen of 4X 250B
<u>Power supply</u>			
G	UCLRL	6 V 5 A Heater current supply reg. p. s.	
H	UCLRL	20V 25A 3X6992	
Interference filter	Spectrolab, Los Angeles	6143-Å	Selects 6143-Å from the neon lamp
Polarizer	Polaroid Inc. , Los Angeles	SM-38	Provides linear polarization of the light at 6143-Å

Table II. (continued)

Item	Manufacturer	Model	Function
L_1 , L_2 and L_3	Edmunds Scientific Co., Barrington, N. J.	----	To concentrate lamp light only
Photomultiplier	RCA Camden, N. J.	7102	Sensing element for light detection
CRO I	Tektronik, Portland, Ore.	541 A	For visible and photographic detection
CRO II	Dumont Lab Inc., Pomona, N. J.	304 H	To monitor collector current.
Camera	Land Polaroid		For photographing oscilloscope traces.
Phase-sensitive detector	Princeton Applied Research, Princeton, N. J.		For noise-free detection; used for plotting excitation function and polarization ratio.
A-C coupled preamplifier	Tektronik, Portland, Ore.	123	Signal amplification
Optical bench	Central Scientific Co., Chicago, Ill.	2m	For mounting interference lenses, and polarizer.

MUB-1384

Fig. 10. Schematic diagram for neon lamp, optical, and detection systems.

The resonance width of the 6143-Å line from the lamp was measured on the photographic plate of the spectra taken using a high-resolution grating.

C. Electron-Gun Power Supplies

Standard commercially available power supplies are used for the electron gun. The grid bias of the electron gun is supplied in the same way as the standard triode amplifier is supplied. Hence, the electron gun itself represents a tetrode vacuum tube. We obtain characteristics of the gun in the same way as in the usual electronic vacuum tube. From this, we can determine the electron current under any experimental condition, if the plate, screen, and grid voltage are known. This is important because, under actual experimental conditions, the collector current one measures is not the current of the electrons because of the formation of ions inside the gun. To make sure that we were able to determine the correct current density, we measured the grid potential with the oscilloscope and the collector and screen potential with a VTVM. Figure 11 shows the electrical connections of the electron gun. Table III contains the essential equipment shown in Fig. 11.

D. Vacuum System

Our vacuum system is completely bakable. Because of this, pressures on the order of 10^{-8} mm of mercury were easily obtained without intensive baking. However, when the electron-gun valve is closed and the heater current is supplied to the cathode heater, the pressure of the gun increased very rapidly. For this reason, we did not run the experiment too long with the same neon sample. Thus, it was necessary to have a fast pump to reduce the waiting period. This required valves having a large diameter. Because of the rather high prices of such valves, we have converted nonbakable Veeco valves to the bakable type by replacing the teflon gasket with a low melting-point indium disk. Two such valves, each having a diameter of 1/2 in.,

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Fig. 11. Schematic diagram of the electron-gun electronics.

are connected to the pumpout tube of the electron gun, and to the diffusion pump via a liquid nitrogen trap backed by a mechanical pump. Because of the geometry involved, it was not possible to enclose the entire system in the oven for baking. We designed the oven to cover the electron gun and the liquid-nitrogen trap. All other parts are baked by heating tapes at about 450° F. A schematic diagram of the vacuum system is shown in Fig. 12. Table IV lists the essential equipment used with vacuum system.

Table III. Table of essential equipment associated with electron gun electronics

Item	Manufacturer	Model	Function
Power supply I	Power Designs Inc., Richmond Hill, N. J.	5015	Provides variable voltage for collector plate.
Power supply II	Power Designs Inc., Richmond Hill, N. J.	5015	Provides variable voltage for screen plate.
Power supply III	Hewlett Packard, Palo Alto, Calif.	712 A	Provides bias for control grid.
Power supply IV	Electro Products Laboratories, Products Inc., Chicago, Ill.	Filtered dc power supply	Power supply for heater.
Square wave generator	Tektronik, Portland, Ore.	105	Provides pulsing potential for control grid.
Cathode ray oscilloscope I	Tektronik Portland, Ore.	541 A	Monitors the potential of the grid.
Cathod ray oscilloscope II	Dumont Lab Inc., Pomona, N. J.	304 H	Monitors the collector current and tests the response of the gun.
Phillip's cathode	Semicon of Calif., Watsonville, Calif.	3/4-in. × 1-in. =OD×L	Source of electrons.
VTVM	Triplet electronic Instr.	850	Voltage measurement.

Fig. 12. Schematic diagram of vacuum system.

Table IV. Table of essential equipment
for vacuum system

Item	Manufacturer	Model	Function
Mechanical pump I	W. N. Welch Manufacturing Co., Chicago, Ill.	Duo-seal vacuum pump	Diffusion pumps backing
Mechanical	W. N. Welch Manufacturing Co., Chicago, Ill.	Duo-seal vacuum pump	To provide rough pump-out without going through the electron gun in order to avoid possible poisoning of the gun.
Diffusion pump	Consolidated Electrodynamics Corp., Rochester, N. Y.	MCF300	Provides high-vacuum pumping.
High temperature bakable valves V ₁ and V ₂	Vacuum Electronic Mfg. Corp., New Hyde Park, N. Y.	1/2 in. ss	These modified valves include a low melting indium disk insert to replace the teflon gasket.
Bakable valves V ₃ , V ₄ , and V ₅	Hoke, Inc., Cresskill, N. J.	XC9385	Isolates part of the system.
V ₆ High vacuum stop coke	Dow Corning		Equalizes the pressure of both columns of the U tube
V ₇ and V ₈ High vacuum stop coke	Dow Corning		Isolates part of the system.

Table IV. (continued)

Item	Manufacturer	Model	Function
Ion gauge tube	Vacuum Electronic Mfg. Corp., New Hyde Park, N. J.	RG75	Low-pressure sensing device. 10^{-5} to 10^{-9} mm/Hg
Ion gauge supply	UCLRL	iX5514G	Power supply for RG75.
Hasting dual vacuum interlock	UCLRL	5Z4994 -1D	Measure forevac pressure in a range of 1 atom to 1 micron and maintain emergency control over the diffusion pump
Manometer (oil)	UCLRL	----	Measures neon-gas pressure
Annealed copper gasket	UCLRL	----	Bakable seal
Leak detector	Consolidated Engineering Corp., Pasadena, Calif	24-101A	Detect leaks
Miscellaneous			
Neon 1 liter reagent grade	Linde Co; Div. of Union Carbide, Tanaward, N. Y.		Source of neon gas

Table IV. (continued)

Item	Manufacturer	Model	Function
Laboratory jack	Cenco-Lerner, Chicago, Ill.	19089	Support
Aluminum dexion bench	UCLRL	-----	Support

IV. ANALYSIS OF EXPERIMENT

A. Lifetimes of the Metastable State ($2^2P_{3/2} \ 3^2S_{1/2}$)₂ by the Electronic-Impact Method.

Reagent-grade neon gas is used in the experiment as a source. The absorption curves obtained for the metastable state of neon are given in Figs. 13 through 24. From analysis of the relaxation curves we find two lifetimes. These absorption curves show there is an initial fast-decay time and a second slow-decay time. The initial-decay time is measured in the region where the previous experiments¹⁻⁴ were unable to measure because of presence of electrons which destroy the metastable atoms. The analysis of the second lifetime was made at the region where the experiment by Phelps and Molnar⁴ was made and is in reasonable agreement with their result.

The data from Figs. 13 through 24 were processed by a computer program, and the results are displayed in Table V. Since there were no apparent marked effects resulting from changing the light intensity, we have performed a straight-forward averaging process. The computer was used only for the first fast initial-decay curve and the second decay curve was analyzed by means of a best fit of a straight-line method.¹³ Also, since the second lifetime was measured in the region where the absorption of light is very small, errors associated with it are very large.

Since some of the results of lifetime are more valid than others, we have assumed a weighting technique. First we assumed that the standard deviation for any one case is proportional to the error for any point of that particular graph. According to Beer (see ref. 13, p.38) the optimal weighting factor is proportional to the inverse square of the standard deviation. First, we average over the entire pressure range in our experiment. The result of this averaging is

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Fig. 13. The oscilloscope trace of the relaxation curve at the pressure of 1.68 mm of mercury for the light transmission of 85.6% and 100% at 1 msec/div.

ZN-3333

Fig. 14. The oscilloscope trace of the relaxation curve at the pressure of 1.68 mm of mercury for the light transmission of 50% and 71.5% at 1 msec/div.

ZN-3340

Fig. 15. The oscilloscope trace of the relaxation curve at the pressure of 1,68 mm of mercury for the light transmission of 15% and 32.6% at 1 msec/div.

ZN-3338

Fig. 16. The oscilloscope trace of the relaxation curve at the pressure of 1.03 mm of mercury for the light transmission of 86.6% and 100% at 1 msec/div.

ZN-3336

Fig. 17. The oscilloscope trace of the relaxation curve at the pressure of 1.03 mm of mercury for the light transmission of 50% and 71.5% at 1 msec/div.

ZN-3334

Fig. 18. The oscilloscope trace of the relaxation curve at the pressure of 1.03 mm of mercury for the light transmission of 15% and 32.6% at 1 msec/div.

ZN-3341

Fig. 19. The oscilloscope trace of the relaxation curve at the pressure of 0.69 mm of mercury for the light transmission of 85.6% and 100% at 1 msec/div.

ZN-3339

Fig. 20. The oscilloscope trace of the relaxation curve at the pressure of 0.69 mm of mercury for the light transmission of 50% and 71.5% at 1 msec/div.

ZN-3335

Fig. 21. The oscilloscope trace of the relaxation curve at the pressure of 0.69 mm of mercury for the light transmission of 15% and 32.6% at 1 msec/div.

ZN-3337

Fig. 22. The oscilloscope trace of the relaxation curve at the pressure of 0.41 mm of mercury for the light transmission of 85.6% and 100% at 1 msec/div.

ZN-3342

Fig. 23. The oscilloscope trace of the relaxation curve at the pressure of 0.41 mm of mercury for the light transmission of 50% and 71.5% at 1 msec/div.

ZN-3343

Fig. 24. The oscilloscope trace of the relaxation curve at the pressure of 0.41 mm of mercury for the light transmission of 15% and 32.6% at 1 msec/div.

Table V. Computer data from Figs. 13 through 24.

Item	$(\tau_{1/2})_1$ (60 min)	$(\tau_{1/2})_1$ (msec)	$\chi^2_{\min}$	% error per point	$\frac{1}{\omega^2} = \frac{1}{\%}$	ω	$\omega(\tau_{1/2})_1$	$\frac{\delta}{\tau - \tau_1}$	δ^2	$\omega\delta^2$
01	0.62	0.37	0.62	08	1.25	1.56	0.58	0.11	0.0121	0.0189
02	0.74	0.44	0.05	10	1.0	1.00	0.44	0.04	0.0016	0.0016
03	0.76	0.46	0.22	14	0.71	0.50	0.23	0.02	0.0004	0.0002
04	0.71	0.43	0.07	21	0.48	0.23	0.10	0.05	0.0025	0.0006
05	0.86	0.52	0.12	12	0.83	0.69	0.36	-0.04	0.0016	0.0011
06	0.72	0.43	0.12	14	0.71	0.50	0.22	0.05	0.0025	0.0013
07	0.80	0.48	0.36	04	2.5	6.25	3.00	0.00		
08	0.70	0.42	0.55	05	2.0	4.00	1.68	0.06	0.0036	0.0144
09	0.78	0.47	1.23	06	1.67	2.79	1.31	0.01	0.0001	0.0003
10	0.64	0.38	0.55	10	1.0	1.00	0.38	0.10	0.0100	0.0100
11	0.73	0.44	0.10	11	0.91	0.83	0.37	0.04	0.0016	0.0013
12	0.70	0.42	0.06	13	0.77	0.59	0.25	0.06	0.0036	0.0021
13	0.82	0.49	1.86	02	5.0	25.00	12.25	-0.01	0.0001	0.0025
14	0.84	0.50	0.63	03	3.33	11.11	5.55	-0.02	0.0004	0.0044
15	0.88	0.53	2.90	03	3.33	11.11	5.89	-0.05	0.0025	0.0278
16	0.76	0.46	1.53	05	2.0	4.00	1.84	0.02	0.0004	0.0016
17	0.84	0.50	1.43	04	2.5	6.25	3.13	-0.02	0.0004	0.0025
18	0.80	0.48	0.26	05	2.0	4.00	1.92	0.00		
19	0.68	0.41	1.34	04	2.5	6.25	2.56	0.07	0.0049	0.0306
20	0.75	0.45	0.38	05	2.0	4.00	1.80	0.03	0.0009	0.0036
21	0.84	0.50	0.13	05	2.0	4.00	2.00	-0.02	0.0004	0.0016
22	0.80	0.48	0.21	07	1.43	2.04	0.98	0.00		
23	0.80	0.48	0.49	08	1.25	1.56	0.75	0.00		
24	0.72	0.43	0.52	09	1.11	1.23	0.53	0.05	0.0025	0.0031
c										
$(\overline{\tau}_{1/2})_1 = \frac{\sum_{i=1}^{24} \omega_i (\tau_{1/2})_{i1}}{\sum_{i=1}^{24}} = \frac{48.12}{100.49} = 0.479 \text{ msec at } 300^\circ\text{K}$						Σ	Σ	Σ		
						=100.49	=48.12	=0.1295		

$$(\bar{\tau}_{1/2})_1 = \frac{\sum_{i=1}^{24} \omega_i (\tau_{1/2})_{1i}}{\sum_{i=1}^{24} \omega_i} = 0.479 \text{ msec}$$

and the standard deviation is

$$\sigma = \left(\frac{\sum_{i=1}^{24} \omega_i \delta_i^2}{\sum_{i=1}^{24} \omega_i} \right)^{1/2} = 0.036 \text{ msec.}$$

So we think it is safe to quote

$$(\bar{\tau}_{1/2})_1 = 0.48 \pm 0.04 \text{ msec at } 300^\circ \text{ K.}$$

Next we analyzed the half-lives for the four different pressure settings. These quantities are tabulated in Table VI, VII, VIII, and IX. The pressure dependence of the half-life is plotted and is shown in Fig. 25.

The second lifetime is obtained by means of the best fit of a straight line (see Fig. 26). For a straight line defined by equation

$$y = a + bx$$

it can be shown¹³ that the desired a and b are obtained by

$$a = \frac{\sum x_n^2 \sum y_n - \sum x_n \sum (x_n y_n)}{k \sum x_n^2 - (\sum x_n)^2},$$

and

$$b = \frac{k \sum (x_n y_n) - \sum x_n \sum y_n}{k \sum x_n^2 - (\sum x_n)^2}.$$

With an assumption that the density of the metastable state decays exponentially as

$$N(t) = N(0) \exp(-t/\tau),$$

we have, taking logarithms of both sides of the above expression,

$$\ln N(t) = \ln N(0) - (1/\tau)t.$$

Table VI. Computer data for pressure of 0.41-mm Hg.

Item	$(\tau_{1/2})_1$ (msec)	$\chi^2_{\min}$	% error per point	$\omega^{\frac{1}{2}}$	ω	$\omega(\tau_{1/2})_1$	δ	δ^2	$\omega\delta^2$
01	0.37	0.62	08	1.25	1.56	0.58	0.06	0.0036	2.16×10^{-4}
02	0.44	0.05	10	1.00	1.00	0.44	0.01	0.0001	0.01×10^{-4}
03	0.46	0.22	14	0.71	0.50	0.23	0.03	0.0009	0.27×10^{-4}
04	0.43	0.07	21	0.48	0.23	0.10	0.00	0.0000	0.00
05	0.52	0.12	12	0.83	0.69	0.36	0.09	0.0081	7.3×10^{-4}
06	0.43	0.12	14	0.71	0.50	0.22	0.00	0.0000	0.00
				Σ =4.48	Σ =1.918			Σ =9.74 $\times 10^{-4}$	

We have pressure = 0.41-mm Hg,

$$(\bar{\tau}_{1/2})_1 = \frac{\sum_{i=1}^6 \omega_i (\tau_{1/2})_{1i}}{\sum \omega_i} = \frac{1.918}{4.48} = 0.428 \text{ msec},$$

and

$$\sigma = \sqrt{\frac{\sum \omega_i \delta_i^2}{\sum \omega_i - 1}} = \sqrt{\frac{9.74 \times 10^{-4}}{4.48 - 1}} = 0.017 \text{ msec}.$$

Therefore,

$$(\bar{\tau}_{1/2})_1 = 0.428 \pm 0.017 \text{ msec at } 300^\circ \text{K},$$

where

$\omega \equiv$ weighting factor,

and

$\delta = \bar{\tau} - \tau_i \equiv$ deviation from the mean.

Table VII. Computer data for pressure of 0.69-mm Hg.

Item	$(\tau_{1/2})_1$ (msec)	$\chi_{\min}^2$	% error per point	$\omega^{\frac{1}{2}}$	ω	$\omega(\tau_{1/2})_1$	δ	δ^2	$\omega\delta^2$
07	0.48	0.36	04	2.5	6.25	3.00	0.03	0.9×10^{-3}	5.6×10^{-3}
08	0.42	0.55	05	2.0	4.00	1.68	0.03	0.9×10^{-3}	3.6×10^{-3}
09	0.47	1.23	06	1.67	2.79	1.31	0.02	0.4×10^{-3}	1.1×10^{-3}
10	0.38	0.55	10	1.0	1.00	0.38	0.07	4.9×10^{-3}	4.9×10^{-3}
11	0.44	0.10	11	0.91	0.83	0.37	0.01	0.1×10^{-3}	0.08×10^{-3}
12	0.42	0.06	13	0.77	0.59	0.25	0.03	0.9×10^{-3}	0.53×10^{-3}
				Σ =15.46		Σ =6.99	Σ =15.81 $\times 10^{-3}$		

We have pressure = 0.69-mm Hg,

$$(\bar{\tau}_{1/2})_1 = \frac{\Sigma \omega_i (\tau_{1/2})_{1i}}{\Sigma \omega_i} = \frac{6.99}{15.46} = 0.452 \text{ msec},$$

and

$$\sigma = \sqrt{\frac{\Sigma \omega_i \delta_i^2}{\Sigma \omega_i - 1}} = \sqrt{\frac{1.58 \times 10^{-3}}{15.46 - 1}} = 3.3 \times 10^{-2}.$$

Therefore,

$$(\tau_{1/2})_1 = 0.452 \pm 0.033 \text{ msec at } 300^\circ \text{ K.}$$

Table VIII. Computer data for pressure of 1.03-mm Hg.

Item	$(\tau_{1/2})_1$ (msec)	$\chi^2_{\min}$	% error per point	$\frac{1}{\omega^2}$	ω	$\omega(\tau_{1/2})_1$	δ	δ^2	$\omega\delta^2$
13	0.49	1.86	02	5.0	25	12.25	0.01	0.1×10^{-3}	2.5×10^{-3}
14	0.50	0.63	03	3.33	11.11	5.55	0.00	0.0	0.0
15	0.53	2.90	03	3.33	11.11	5.89	0.03	0.9×10^{-3}	9.99×10^{-3}
16	0.46	1.53	05	2.00	4.00	1.84	0.04	0.4×10^{-3}	1.6×10^{-3}
17	0.50	1.43	04	2.50	6.25	3.13	0.00	0.00	0.0
18	0.48	0.26	05	2.0	4.00	1.92	0.02	0.4×10^{-3}	1.6×10^{-3}
				Σ	Σ	Σ			
				=61.47	=30.58				
							Σ		
							15.69×10^{-3}		

We have pressure = 1.03-mm Hg,

$$(\bar{\tau}_{1/2})_1 = \frac{\Sigma \omega_i (\tau_{1/2})_{1i}}{\Sigma \omega_i} = \frac{30.58}{61.47} = 0.497 \text{ msec,}$$

and

$$\sigma = \sqrt{\frac{\Sigma \omega_i \delta_i^2}{\Sigma \omega_i - 1}} = \sqrt{\frac{15.69 \times 10^{-3}}{6.047 \times 10}} = 1.6 \times 10^{-2} \text{ msec.}$$

Therefore,

$$(\bar{\tau}_{1/2})_1 = 0.497 \pm 0.016 \text{ msec at } 300^\circ \text{K.}$$

Table IX. Computer data for pressure of 1.68-mm Hg.

Item	$(\tau_{1/2})_1$ (msec)	$\chi^2_{\min}$	% error per point	ω^2	ω	$\omega(\tau_{1/2})_1$	δ	δ^2	$\omega\delta^2$
19	0.41	1.34	04	2.5	6.25	2.56	0.04	1.6×10^{-3}	10.0×10^{-3}
20	0.45	0.38	05	2.0	4.00	1.80	0.00	0.0	0.0
21	0.50	0.13	05	2.0	4.00	2.00	0.05	2.5×10^{-3}	10.0×10^{-3}
22	0.48	0.21	07	1.43	2.04	0.98	0.03	0.9×10^{-3}	1.8×10^{-3}
23	0.48	0.49	08	1.25	1.56	0.75	0.03	0.9×10^{-3}	1.4×10^{-3}
24	0.43	0.52	09	1.11	1.23	0.53	0.02	0.4×10^{-3}	0.49×10^{-3}
				Σ	Σ				Σ
					=19.08	=8.62			=23.69 $\times 10^{-3}$

We have pressure = 1.68-mm Hg,

$$(\bar{\tau}_{1/2})_1 = \frac{\Sigma \omega_i (\tau_{1/2})_{1i}}{\Sigma \omega_i} = \frac{8.62}{19.08} = 0.451 \text{ msec.}$$

and

$$\sigma = \sqrt{\frac{\Sigma \omega_i \delta_i^2}{\Sigma \omega_i - 1}} = \sqrt{\frac{23.69 \times 10^{-3}}{18.08}} = 3.6 \times 10^{-2} \text{ msec.}$$

Therefore,

$$(\bar{\tau}_{1/2})_1 = 0.451 \pm 0.036 \text{ msec at } 300^\circ \text{K.}$$

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Fig. 25. The pressure dependence of the half-life $(\tau_{1/2})_1$ at 300°K.

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Fig. 26. Plot of the decay curve to show the existence two lifetimes for $p=0.69$ mm Hg at 300°K .

This is put in the desired form by placing $\ln N(t) = y$, $\ln N(0) = a$, $(1/\tau) = -b$, and $t = x$. Points for the best fit of a straight line are taken from the data in Figs. 13 through 24 for the time region of 2 to 4 msec for the second relaxation times. Details of the method of calculation for the best fit of a straight line are given in Beer.¹³ The results we obtained are tabulated in Table X.

B. Excitation Function

The measured value of the excitation function $\Delta I/I$ is shown in Figs. 27 through 30 for the pressures of 0.41, 0.69, 1.03, and 1.69 mm of Hg. Since we are also interested in the polarization ratio, we linearly polarize the incident light parallel to and perpendicular to the direction of the electron beam. Let $\Delta I_{||}$ be the amount of the absorption when the linear polarization is parallel to the direction of the electron beam, and let $\Delta I_{\perp}$ be the absorption when the linear polarization is perpendicular to the direction of the electron beam. Then the net absorption is

$$\Delta I = \Delta I_{||} + \Delta I_{\perp} . \quad (23)$$

Let the intensity of the incident light I be composed of two components $I_{||}$ and $I_{\perp}$. Then the intensity I is

$$I = I_{||} + I_{\perp} . \quad (23')$$

The light source is unpolarized. Therefore, $I_{||}$ should be equal to $I_{\perp}$. However, the response of the monochrometer used is different for the two orientations of the polarization of the light. We must take this into account for the evaluation of the excitation function. First we measured $I_{||}$ and $I_{\perp}$ in the absence of absorption. This gives us the characteristics of the response of the monochrometer used with respect to the orientation of the polarization of the light. Let $\epsilon = \frac{I_{||}}{I_{\perp}}$. In terms of this, the incident intensity I becomes

$$I = I_{||} + \epsilon I_{\perp} , \quad (23)''$$

and the total absorption ΔI becomes

$$\Delta I = \Delta I_{||} + \epsilon \Delta I_{\perp} . \quad (23)'''$$

Table X. Values of $(\tau_{1/2})_2$.

Half-life $(\tau_{1/2})_2$ in msec	Pressure (mm Hg) at 300°K
2.5 (1.8)	1.68
4.8 (4.0)	1.03
8.3 (7.5)	0.69
$(\tau_{1/2})_2 > 8.3$	0.41

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Fig. 27. Excitation function of the neon metastable state using 6143 Å optical absorption line at a pressure of 0.41 mm of Hg at 300°K.

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Fig. 28. Excitation function of the neon metastable state using 6143 Å optical absorption line at a pressure of 0.69 mm of Hg at 300° K.

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Fig. 29. Excitation function of the neon metastable state using 6143 Å optical absorption line at a pressure of 1.03 mm of Hg at 300° K.

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Fig. 30. Excitation function of the neon metastable state using 6143 Å optical absorption line at a pressure of 1.68 mm of Hg at 300°K.

Therefore,

$$\frac{\Delta I}{I} = \frac{\text{Absorbed Intensity}}{\text{Incident Intensity}} = \frac{\Delta I_{||} + \epsilon \Delta I_{\perp}}{I_{||} + \epsilon I_{\perp}} \quad (24)$$

Of course, if we take the ratio $I_{\perp} / I_{||}$ instead of $I_{||} / I_{\perp}$ the quantity in Eq. (23)" and (23)'" will vary. However, since we are taking a ratio in Eq. (24), $\Delta I / I$ will not change. The maximum value for $\Delta I / I$ is about 2%. This is consistent with the experimental value obtained by Milats and Orenstein.⁵

A phase-sensitive detector preceded by an amplifier is used in order to obtain less noisy detection of the signal. We have calibrated the amplification factor of the amplifier and the meter readings of the phase-sensitive detector against known signal sources. The amplifier was used before the signal was fed into the phase-sensitive detector since we noticed that the fluctuations of the phase sensitive detector were less when the signal was first amplified.

C. Half-Width Measurement of the 6143-Å Line

The dispersion of the grating used in measuring the width of the resonance line 6143-Å was determined by exposing four lines of neon 6163.59-Å, 6143.06-Å, 6128.45-Å, and 6096.16-Å on a plate and measuring the separation between them. The mean average dispersion for these lines was 0.485-Å/mm. Knowing the scanning speed of the densitometer, we calibrated the recorder paper of the densitometer by measuring the speed of the recorder paper. The calibration of the paper turned out to be 2mm of the plate per 12 in. of the recorder paper. Therefore, we get the calibration of the recorder paper from the dispersion to be

$$\frac{2\text{mm}}{12 \text{ in}} \times \frac{0.485\text{-}\text{\AA}}{1 \text{ mm}} = 0.0884\text{-}\text{\AA}/\text{in.}$$

We exposed the neon lamp light from the spectroscope on the plate for 1 sec, 2 sec, 4 sec, and 9 sec. The densitometer curves corresponding to these exposure times are given in Figs. 31 through 34. Since the photographic plate has logarithmic response, the amplitude obtained from the densitometer curve is proportional to $\log(\text{intensity})$. We assumed that the energy of the light that exposes the photographic plate is linearly proportional to the amount of time exposure. Therefore, if we plot the density vs $\log(\text{time exposure})$ we should expect a straight line (see Fig. 35). If there is a deviation from the straight line, this corresponds to either extreme under exposure or saturation of the photographic plate. Therefore, to obtain the half-width of the resonance line, we should use the point of the densitometer curve which lies in the linear region of the density vs $\ln(I)$ curve. The density corresponding to the half-height is, therefore, $\ln(1/2I)$. The intensity vs $\ln(I)$ curve is given in Fig. 33. We shall use the one with the 4-sec exposure time since this curve lies approximately in the middle of the graph in Fig. 33. The maximum density of the 4-sec exposure curve is 64%. The density corresponding the half-width is 38% as seen from the graph in Fig. 33. The density used in Fig. 33 is measured in percentage with respect to the height in the densitometer curve corresponding to the total darkness. Thus we get the half-width of the resonance line to be 0.45 inches from the densitometer trace in Fig. 32. This corresponds to the half-width of $0.0364\text{-}\text{\AA}$ for the $6143\text{-}\text{\AA}$ line. We feel that it is safe to assign an error of 15%. *

*Private conversation with Professor Sumner P. Davis.

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Fig. 31. Densitometer curve for exposure time of 1 second.
The curve at right corresponds to 100% absorption.

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Fig. 32. Densitometer curve for exposure time of 2 seconds.
The curve at right corresponds to 100% absorption.

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Fig. 33. Densitometer curve for exposure time of 4 seconds.
The curve at right corresponds to 100% absorption.

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Fig. 34. Densitometer curve for exposure time of 9 seconds.
The curve at right corresponds to 100% absorption.

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Fig. 35. Graph of density vs log (time exposure).

Therefore, we obtain

$$\Delta\lambda = 0.036 \pm 0.006 \text{ -}\text{\AA} \text{ ,}$$

which corresponds to

$$\Delta\nu = 2880 \pm 450 \text{ Mc/sec.}$$

With the values we obtained for the excitation function, lifetimes, and the half-width of the resonance radiation, we are now in a position to determine the density of the metastable state using Eq. (15).

D. Density Measurement of the Metastable Neon Atoms

We assume that the metastable state atoms will be destroyed when they collide with other atoms. Since the mean free path of the atom at 1 mm of mercury pressure is less than 1 mm, and because the metastable atoms will scatter isotropically after they are produced by the electron beam, we shall assume that the optical absorption length is the diameter of the electron beam. With this assumption

$$N = \frac{1}{\left(\frac{l}{\Delta\nu}\right)\left(\frac{\pi e^2}{mc}\right)|f_{ij}|} \times \left(\frac{\Delta I}{I}\right) = 2.88 \times 10^{11} \times \left(\frac{\Delta I}{I}\right) \quad (25)$$

with $f_{ij} = 0.22$.

We believe that the main source of uncertainty in calculating the density arises from the error associated with the measurement of the half-width of the resonance radiation and with the calculation of the oscillator strength f_{ij} . To obtain the density of the metastable state, we simply multiply $(\Delta I/I)$, expressed in percentage in Figs. 27 through 30, by 2.88×10^9 . Table XI contains the maximum density for the four pressure settings under the electronic-impact condition.

Table XI. Density of the metastable state ($2^3P_{1/2} 3^2S_{1/2}$)₂.

Maximum density of the metastable state ($\times 10^9$ atoms/cc)	Electron energy (V)	Pressure (mm Hg)
6.3(1.8)	23	0.41
5.3(1.5)	23	0.69
5.8(1.8)	19	1.03
3.7(1.2)	19	1.68

E. Total Excitation Cross Section

The total excitation cross section is determined by using Eq. (19), where

$$\sigma_t = \frac{1}{N_0(J/e)} \times \frac{1}{(\ell/\Delta\nu)(\pi e^2/mc) f_{ij} \tau} \times \frac{\Delta I}{I}.$$

With the value $\ell = 3/4$ in., $\Delta\nu = 2880$ Mc/sec and $f_{ij} = 0.22$, we may write Eq. (19) as

$$\sigma_t = 2.88 \times 10^{11} \times \frac{1}{N_0(J/e) \tau} \times \frac{\Delta I}{I}. \quad (26).$$

The total cross section for electronic-impact excitation may be obtained by simply multiplying the excitation functions in Figs. 27 through 30 by $2.88 \times 10^9 / N_0(J/e) \tau$ where N_0 is the number of ground-state atoms/cm³ and (J/e) is the electron flux since $(\Delta I/I)$ is expressed in percentage in these figures. However, we shall tabulate the maximum values of the total cross sections for the four pressure settings in Table XII. The details of the calculation are given in Appendix III.

F. Polarization Ratio

The polarization ratio defined by the relation $\frac{\Delta I_{\perp} - \Delta I_{\parallel}}{\Delta I_{\parallel} + \Delta I_{\perp}}$

is given in Table XIII as a function of excitation energy. As Table XIII shows, the polarization ratio has a positive sign very near the excitation threshold. The threshold of excitation for the metastable state is 16.5 eV and the sign of the polarization ratio is positive only for the excitation energy of 17 and 18 V. Unfortunately, this is the region where the absorption is about to occur. The reason why we should have a positive sign can be explained in the following way. Figures 36 and 37 show the relevant transitions with corresponding relative-transition probabilities.

Table XII. Cross section for excitation.

Maximum cross section ($\times 10^{-19} \text{ cm}^2$)	Electron energy (V)	Pressure (mm Hg)
1.54(0.5)	23	0.41
0.63(0.2)	23	0.69
0.48(0.2)	19	1.03
1.95(0.6)	19	1.68

Table XIII. Polarization ratio.

Polarization ratio $(\Delta I_{\perp} - \Delta I_{\parallel})/(\Delta I_{\perp} + \Delta I_{\parallel})$ %	Pressure (mm Hg)	Excitation voltage
-11(26)	0.41	19
-2.2(9)	0.41	21
-1.4(3)	0.41	22
+18(20)	0.69	17
-9(4)	0.69	19
-5.8(3)	0.69	20
-11.5(5)	0.69	21
+3.5(20)	1.03	17
+0.2(3)	1.03	18
-0.8(2)	1.03	19
-1.7(3)	1.03	20
-0.8(5)	1.03	21
+6.5(18)	1.63	17
+1.9(2)	1.63	18
-1.6(1)	1.63	19
-4.0(2)	1.63	20
-0.8(2)	1.63	21

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Fig. 36. Relevant relative transition probabilities when the electric vector of the incident light is parallel to the direction of the electron beam.

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Fig. 37. Relevant transition probabilities when the electric vector of the incident radiation is perpendicular to the direction of the electron beam.

These transition probabilities are calculated by noting that the electron-dipole operator has the same transformation properties as the irreducible tensor operator of rank one, as mentioned in Sec. II, so that

$$|\langle i | \underline{p} | j \rangle|^2 = \sum_{M_J} \sum_{M_{J'}} \sum_q \begin{pmatrix} J & 1 & J \\ -M_J & q & M_{J'} \end{pmatrix}^2 |\langle i || \underline{p} || j \rangle|^2.$$

Therefore, the relative-transition probability is proportional to the square of the 3-j symbol. For the case of the electric vector of the incident light parallel to the direction of the electron beam, the optical-dipole selection rule $\Delta M=0$ holds true. Hence, the contribution for the relative transition probabilities comes from

$$\begin{pmatrix} 2 & 1 & 2 \\ -2 & 0 & 2 \end{pmatrix}^2 = \frac{4}{30}, \quad \begin{pmatrix} 2 & 1 & 2 \\ -1 & 0 & 1 \end{pmatrix}^2 = \frac{1}{30}, \quad \begin{pmatrix} 2 & 1 & 2 \\ 0 & 0 & 0 \end{pmatrix}^2 = 0,$$

and

$$\begin{pmatrix} 2 & 1 & 2 \\ 2 & 0 & -2 \end{pmatrix}^2 = \frac{4}{30}.$$

When the electric vector is perpendicular to the direction of the electron beam, the contribution for the relative-transition probabilities comes from

$$\begin{aligned} \frac{1}{2} \begin{pmatrix} 2 & 1 & 2 \\ -2 & 1 & 1 \end{pmatrix}^2 &= \frac{1}{2} \begin{pmatrix} 2 & 1 & 2 \\ 1 & 1 & -2 \end{pmatrix}^2 = \frac{1}{30}, \quad \frac{1}{2} \begin{pmatrix} 2 & 1 & 2 \\ -1 & -1 & 2 \end{pmatrix}^2 \\ &= \frac{1}{2} \begin{pmatrix} 2 & 1 & 2 \\ 2 & -1 & -1 \end{pmatrix}^2 = \frac{1}{30}, \end{aligned}$$

$$\frac{1}{2} \begin{pmatrix} 2 & 1 & 2 \\ -1 & 1 & 0 \end{pmatrix}^2 = \frac{1}{2} \begin{pmatrix} 2 & 1 & 2 \\ 1 & -1 & 0 \end{pmatrix}^2 = \frac{3}{60}, \quad \text{and}$$

$$\frac{1}{2} \begin{pmatrix} 2 & 1 & 2 \\ 0 & 1 & -1 \end{pmatrix}^2 = \frac{1}{2} \begin{pmatrix} 2 & 1 & 2 \\ 0 & -1 & 1 \end{pmatrix}^2 = \frac{3}{60},$$

since the optical-dipole selection rule $\Delta M = \pm 1$. Using the relative-transition probabilities derived above, we obtain the ratio between the transmitted intensities $\Delta I_{\perp}$ and $\Delta I_{\parallel}$,

$$\frac{\Delta I_{\perp}}{\Delta I_{\parallel}} = \frac{8N_2 + 2N_1}{2N_2 + 5N_1 + 3N_0}, \quad (27)$$

where N_2 , N_1 , and N_0 correspond to the population of the magnetic sublevel with $M_J = \pm 2$, ± 1 , and 0 , with an assumption that the population corresponding to the magnetic sublevel with $M = M_J$ is equal to that with $M = -M_J$. Therefore, in terms of these population distributions, the polarization ratio becomes

$$\frac{(2N_2 + 5N_1 + 3N_0) - (8N_2 + 2N_1)}{(2N_2 + 5N_1 + 3N_0) + (8N_2 + 2N_1)} = \frac{-6N_2 + 3N_1 + 3N_0}{10N_2 + 7N_1 + 3N_0}. \quad (28)$$

From Eq. (29), we can clearly see that when there is an equilibrium in the population distribution, i. e. when $N_2 = N_1 = N_0$, the polarization ratio vanishes. On the other hand, if there is a preferential population of the magnetic sublevels $M_J = 0$ and, because of electron exchange, also of $M_J = \pm 1$, we see that the polarization ratio must be positive. This is the quantity we see from the measurement of the polarization ratio. Since the polarization ratio is positive at the excitation energy very close to the threshold of excitation, we are certain that we are producing the preferential population of the magnetic sublevels. This effect may be qualitatively explained from the idealized model.¹⁴

→ Consider the case where an electron possesses a kinetic energy just sufficient to excite the atom to the metastable state. The angular momentum of the electron before impact is in a plane normal to the direction of propagation. The speed of the electron after the impact is zero and it cannot have any angular momentum except in the spin coordinate. From a consideration of the conservation of the angular momentum neglecting the spin, the impact has transferred an angular momentum perpendicular to the direction of the propagation to the atom.

The transition from the ground state to the excited state cannot be accompanied by a change of the angular-momentum component parallel to the direction of propagation, and the transition is made with $\Delta M_J=0$. This explains quantitatively the reason for the preferential population of the magnetic sublevel with $M_J=0$. More quantitative explanations are made by Bethe¹⁵ and Oppenheimer.¹⁶ The probability of excitation of the atomic level indicated by index n is given by

$$F(\underline{p} - \underline{p}') = - \int u_n^* u_0 \sum_j e^{i(\underline{p} - \underline{p}') \cdot \underline{r}_j} d\tau, \quad (29)$$

where u_n and u_0 are eigenfunctions of the excited state n and the initial state, $\underline{p}$ and $\underline{p}'$ are the linear momenta of the incident electron before and after the impact, and $\underline{r}_j$ is the spatial coordinate of the j th electron. The integration is over the configuration space of the atomic electrons. In our case, the transition concerned can be regarded as one involving a single electron only so that we may drop the summation over the atomic electrons. From a consideration of the invariance of the integral with respect to the rotation of the coordinate by an angle ϕ about $\underline{p}-\underline{p}'$ which amounts to the multiplication of the eigenfunction u_n and u_0 by $e^{im\phi}$ and $e^{im_0\phi}$ respectively, one obtains the condition $m_0-m=0$. We may consider the two limiting cases.¹⁵

1. The energy of the incident electron is near the threshold of the excitation of the atomic-energy level. Then we have $\underline{p}' = 0$. Therefore, $\underline{p} - \underline{p}'$ is in the direction of $\underline{p}$. This corresponds to the preferential population with $\Delta M_J=0$. In case this excitation corresponds to that of the optically allowed transition to the ground state with $J = 0$, the emitted light will be linearly polarized parallel to the direction of the incident electron.
2. The energy of the incident electron is much larger than the threshold of the excitation such that $\underline{p} = \underline{p}'$. Then $\underline{p} - \underline{p}'$ is perpendicular to $\underline{p}$. This corresponds to the case where the excited state will have $|M_J| > 0$. For the optically allowed transition, the emitted light will be linearly polarized perpendicular to $\underline{p}$.

So far the effect of the electron-spin exchange is completely ignored. To apply the conservation of angular momentum correctly, we must consider the total spin of the atom plus that of the incident electron. The formula in Eq. (29) requires conservation of the spin of the atomic levels because it ignores the possibility of electron exchange. It can be shown that the spin-exchange effect can cause the excitation of the excited state with $\Delta M_J = \pm 1$.¹⁴

Our experimental studies of polarization ratio partially verified the results of Bethe,¹⁵ since the polarization ratio was positive very near the threshold of excitation and negative at the energy higher than the threshold of excitation by 3V or more.

From Eq. (28) we see that at the excitation energy very close to the threshold of excitation, we obtained a preferential population of N_1 and N_0 , since the polarization ratio was positive. At higher energies, the polarization ratio was negative. This corresponds to the preferential population of N_2 over the population N_1 and N_0 .

The most serious problem we are facing at present is as follows. Because the preferential population takes place at the excitation energy very close to the threshold of excitation, the absorption is very small. We believe this is the reason that we were not able to observe an rf resonance. Since we must live with the fact that the preferential population of N_1 and N_0 takes place very close to the threshold of excitation, the only hope we have seems to be to increase the optical absorption.

V. CONCLUSION

The peak of the excitation function varied from 1.3 to 2.5%. This is in agreement with the experiments of Milatz and Orenstein.⁵ The density of metastable neon atoms ($2^2P_{3/2} S_{1/2}$)₂ they measured was about $5 \times 10^8 / \text{cm}^3$, whereas our measured value is about $5 \times 10^9 / \text{cm}^3$. However, no measurement on the half-width of neon was made by Milatz and Orenstein. They used a calculated value with the assumption that the broadening of the line is due to the Doppler shift. The value they assumed is much smaller than the value we obtained experimentally. By using our experimentally observed value, we obtain a rough agreement in density. The initial rise of the excitation function is very fast; however, after the peak of the excitation function, the decent is much slower than the initial rise. Also, it does not come down to zero. As is explained in detail by Massey and Burhop,¹⁷ we believe that this is due to the excitation of higher states which cascade down to the metastable state because the excitation energy of the electrons is much higher than the threshold of excitation at this range.

The maximum total cross section we measured is about $1/200$ of that corresponding to the area due to the first Bohr orbit for hydrogen atom. This is not too surprising, even though the metastable atom has the principal quantum-number 3 so that its area should be larger, since the transition caused by the electronic impact is that of an optically forbidden transition (see ref. 17, p. 165). No satisfactory theory exists to explain the above effects yet.

We observed that there are two lifetimes one of which is much shorter than the other. The shorter lifetime showed very little change with respect to pressure variations whereas the longer lifetime showed a considerable amount of change. The one with the longer lifetime increase as the pressure gets lower as expected. We believe that the metastable atoms with the shorter lifetime are produced by the electronic excitation directly whereas those with the longer lifetime are

produced by the higher metastable states cascading down to the lower metastable state. At present, we are not certain if the longer lifetimes are produced by the cascading effect from a single state or several states. The upper metastable state ($^2P_{1/2}$ $^2S_{1/2}$)₀ has been measured by Phelps and Molnar.⁴ Unfortunately, because of rather large noise, they were not able to analyze the decay too well.

It is not easy for us to quantitatively explain the mechanism causing the half-life to be of the order of magnitude of 0.54 msec for the shorter lifetime. However, we think that we may be able to explain the mechanism qualitatively in the following way.* There are four closely spaced energy levels as are shown in Fig. 38. Of these, the ones with total angular momentum of $J = 1$ are the radiative states with the electric-dipole transitions to the ground state. However, at a relatively high pressure of neon, these four levels tend to establish a population equilibrium. Thus, the lowest metastable state can decay to the ground state by transferring its energy to the state with $J = 1$ by collision either with ground state atoms or with atoms in those four closely spaced levels. A calculation explaining this effect for the two-level system was made by Bennett:¹⁸ however, when there are four levels involved, the calculation is more complicated.

To observe an rf resonance of the metastable-state atoms produced by electronic impact, it is necessary for the polarization ratio to be as large as possible. No information was available prior to our experiment concerning this. The main reason we measured this condition was to investigate the possibility of observing an rf resonance. We observed that the positive values of the polarization ratio occur only at electronic excitation energies of 17 and 18V. The threshold of excitation is 16.5 eV for the lowest metastable state. This is the region where the optical absorption of the resonance radiation just begins to take place. However, we did obtain the polarization ratio of 18(20)% at the pressure of 0.69-mm mercury. Unfortunately, the signal-to-noise ratio was less than one even with the phase-sensitive detector. This, we believe, is the reason we could not observe the rf resonance.

*Private conversation with Dr. W. R. Bennett.

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Fig. 38. Relative energy spacing of the lowest excited electron configuration of the noble gas atom.

Since, the absorption of light is linearly proportional to the optical path length for a small coefficient of absorption, the next obvious thing to do seems to be to construct an electron gun with a rectangular cathode of much longer length. Additionally, we are investigating the possibility of causing the multiple reflection of the incident light between mirrors to increase the optical path length. The signal-to-noise ratio of about 15 seems obtainable with a cathode whose length is about 30 cm even without the multiple reflection of the incident light. Quite recently, this sort of cathode was constructed at Bell Telephone Laboratories, Murray Hill, N. J., by Dr. W. R. Bennett. So it seems that if we can not obtain an rf resonance with one cathode, we may add a few more until we obtain sufficient absorption at a very low pressure of neon gas. The lower density appears necessary to obtain sufficient alignment since the collision of the aligned metastable atoms with other atoms should be reduced. Although it seems simple to construct such an electron gun, we believe that the technical problems associated with such a system are quite large.

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APPENDICES

$$\text{I. Validity of the Approximation } \Delta I/I = \frac{\ell \int_{-\infty}^{\infty} k_{\nu} d\nu}{\Delta \nu}$$

Following Ladenburg and Reiche,¹⁹ we assume

$$I_0(\nu) = C e^{-4(\nu - \nu_D)^2 \ln 2 / (\Delta \nu_D)^2}$$

where $\Delta \nu_D$ is the Doppler breadth of the absorption line. Then we have

$$\Delta I/I = \frac{\ell \int_{-\infty}^{\infty} I_0(\nu) k_{\nu} d\nu}{I_0(\nu) d\nu} = \frac{\ell \int_{-\infty}^{\infty} C e^{-4(\nu - \nu_D)^2 \ln 2 / (\Delta \nu_D)^2} k_{\nu} d\nu}{\int_{-\infty}^{\infty} C e^{-4(\nu - \nu_D)^2 \ln 2 / (\Delta \nu_D)^2} d\nu} \quad (1)$$

Now we obtain

$$\int_{-\infty}^{\infty} C e^{-4(\nu - \nu_D)^2 \ln 2 / (\Delta \nu_D)^2} d\nu = \frac{\sqrt{\pi}}{4\sqrt{\ln 2}} \Delta \nu_D$$

If we assume $k_{\nu} = k_0 e^{-4(\nu - \nu_D)^2 \ln 2 / (\Delta \nu_D)^2}$,

then we get

$$\Delta I/I = \frac{\ell k_0 \frac{1}{\sqrt{2}} \frac{\sqrt{\pi}}{4\sqrt{\ln 2}} \Delta \nu_D}{\frac{\sqrt{\pi}}{4\sqrt{\ln 2}} \Delta \nu_D} = \frac{\ell k_0}{\sqrt{2}} \quad (2)$$

More rigorous treatment by Malinowski,²⁰ Orthmann,²¹ Kunze,²² de Grott,²³ and Zemanski,²⁴ using the method of Ladenburg and Reiche,¹⁹ shows

$$\Delta I/I = \frac{\int_{-\infty}^{\infty} e^{-\frac{4(\nu-\nu_0)^2 \ln^2}{\Delta \nu_D}} \left(1 - e^{-k_0 \ell e^{-\frac{4(\nu-\nu_0)^2 \ln^2}{\Delta \nu_D}}} \right) d\nu}{\int_{-\infty}^{\infty} e^{-\frac{4(\nu-\nu_0)^2 \ln^2}{\Delta \nu_D}} d\nu}$$

$$= \frac{k_0 \ell}{\sqrt{2}} - \frac{(k_0 \ell)^2}{2! \sqrt{3}} + \dots + \frac{(-)^n (k_0 \ell)^n}{n! \sqrt{1+n}} \quad (3)$$

for the case in which the emission line breadth = absorption line breadth. The first term is exactly equal to the case of our approximation

$$\Delta I/I = \frac{\ell \int_{-\infty}^{\infty} k_\nu d\nu}{\Delta \nu}$$

Using the method of Ladenburg and Reiche,¹⁹ Kopfermann and Tietze²⁵ measured the absorption of mercury vapor and found the maximum absorption coefficient to vary with the number of absorbing atoms, N, quite linearly for small values of N according to the law

$$k_0 = 1.34 \times 10^{-13} N.$$

If we assume that neon atoms at low pressure also behave approximately like the mercury atoms, we are well justified in neglecting the higher order term in the expansion of Eq. (3) since the measured value of the metastable atoms is on the order of $10^{-9}/\text{cm}^3$ and $\ell \simeq 2$ cm.

II. Calculation of $\left(\int_0^{\infty} R_{30} R_{31} r dr \right)^2$.

The method used here is that given by Bates and Damgaard.⁸ In the central field model the functions R_i and R_f satisfy a differential equation

$$\frac{d^2 R}{dr^2} + \left(2V - \frac{\ell(\ell+1)}{r^2} - \epsilon \right) R = 0, \quad (1)$$

where V is the potential, ℓ the azimuthal quantum number and ϵ is the energy eigenvalue. If we write

$$\int_0^{\infty} R_i R_f r dr = \int_0^{r_c} R_i R_f r dr + \int_{r_c}^{\infty} R_i R_f r dr, \quad (2)$$

it can be shown that

$$\frac{\int_0^{r_c} R_i R_f r dr}{\int_{r_c}^{\infty} R_i R_f r dr} \approx 1/100$$

for $r_c \approx 2\text{\AA}$ from the origin for the wave functions determined by Fock and Petrashen²⁶ for Na I, and for the wave functions determined by Hartree, Hartree and Swirles,²⁷ and Bates and Damgaard²⁸ for O II. Therefore, the wave function appearing in Eq. (2) need only be accurate at moderate and large radial distances. This is the basis for replacing the potential V by C/r where C is the excess charge on the nucleus when the active electron is removed. Therefore, the differential equation for R becomes

$$\frac{d^2 R}{dr^2} + \left(\frac{2C}{r} - \frac{\ell(\ell+1)}{r^2} - \epsilon \right) R = 0 \quad (3)$$

with the boundary condition

$$R \longrightarrow 0 \text{ as } r \longrightarrow \infty. \quad (4)$$

The solution chosen as most appropriate, given by Eddington²⁹ and Sugira,³⁰ is

$$R = W_{n^*, \ell + \frac{1}{2}} \left(\frac{2Cr}{n^*} \right) \quad (5)$$

with $n^* = C/\epsilon^{1/2}$ and $W_{n^*, \ell + \frac{1}{2}}$ the confluent hypergeometric function defined as the solution of the equation

$$\frac{d^2 W_{n^*, \ell + \frac{1}{2}}}{dr^2} + \left[\frac{2C}{r} - \frac{C^2}{(n^*)^2} - \frac{\ell(\ell+1)}{r^2} \right] W_{n^*, \ell + \frac{1}{2}} = 0. \quad (6)$$

This has the asymptotic expansion

$$a_1 = \frac{n^*}{2C} [\ell(\ell+1) - n^*(n^*-1)] \quad (7)$$

and

$$a_t = a_{t-1} \left\{ \frac{n^*}{2tC} [\ell(\ell+1) - (n^*-t)(n^*-t+1)] \right\}. \quad (8)$$

For the purpose of normalization, the confluent hypergeometric function should be multiplied by the normalization factor

$$1/\left[n^{*2} \Gamma(n^* + \ell - 1) \Gamma(n^* - \ell)/C \right]. \quad (9)$$

Let the value of n^* and ℓ associated with R_i be n^{*i} and ℓ^i , and let those associated with R_f be $n^{*''}$ and ℓ'' . The general terms in the normalized expansion of R_i and R_f may be written in the form

$$\sum_{p'=0}^{\infty} C_{p'}(n^{*'}, l') C^{\frac{1}{2}} (Cr)^{n^{*'}-p'} \exp \left(-\frac{Cr}{n^{*'}} \right), \quad (10)$$

and

$$\sum_{p''=0}^{\infty} C_{p''}(n^{*''}, l'') C^{\frac{1}{2}} (Cr)^{n^{*''}-p''} \exp \left(-\frac{Cr}{n^{*''}} \right) \quad (11)$$

respectively, the coefficients $C_{p'}(n^{*'}, l')$ and $C_{p''}(n^{*''}, l'')$ being known constants. It follows that

$$\begin{aligned} \int_0^{\infty} R_i R_f r dr &= \sum_{p'} \sum_{p''} \int_0^{\infty} C_{p'}(n^{*'}, l') C_{p''}(n^{*''}, l'') (Cr)^{n^{*'}+n^{*''}+1-p'-p''} \\ &\quad \exp \left[-Cr \left(\frac{n^{*'}+n^{*''}}{n^{*'} n^{*''}} \right) \right] dr \quad (12) \\ &= \frac{1}{C} \sum_{p'} \sum_{p''} C_{p'}(n^{*'}, l') C_{p''}(n^{*''}, l'') \left(\frac{n^{*'}+n^{*''}}{n^{*'} n^{*''}} \right)^{n^{*'}+n^{*''}+2-p'-p''} \\ &\quad \cdot \Gamma(n^{*'}+n^{*''}+1-p'-p''). \end{aligned}$$

This series terminates if $n^{*'}$ and $n^{*''}$ are integers, but in general is of the form of an asymptotic expansion. It can, however, be evaluated with sufficient accuracy by ignoring terms for which

$$p' + p'' < n^{*'} + n^{*''} - 1.$$

The computations in the evaluation of series were carried out by Bates and Damgaard⁸ for s-p, p-d, and d-f transitions. The integral

$$\sigma^* = \frac{1}{(4\ell^2 - 1)} \left(\int_0^\infty R_i R_f r dr \right)^2$$

is expressed in compact form

$$\mathcal{F}(n_\ell^*, \ell) = \left[\frac{3n_\ell^*}{2} \left\{ \frac{(n_\ell^{*2} - \ell^2)}{4\ell^2 - 1} \right\}^{1/2} \right]$$

and

$$\mathcal{G}(n_{\ell-1}^*, n_\ell^*, \ell) = \left[\frac{2C}{3n_\ell^*} \frac{1}{(n_\ell^{*2} - \ell^2)} \right] \int_0^\infty R(n_{\ell-1}^*, \ell-1, C) R(n_\ell^*, \ell, C) r dr$$

The function $\mathcal{F}(n_\ell^*, \ell)$ and $\mathcal{G}(n_{\ell-1}^*, n_\ell^*, \ell)$ are tabulated in the table by Bates and Damgaard.

The energy parameter ϵ is taken to be the energy required to remove the electron from the particular level concerned for a single electron outside the closed shell. We applied the same condition to neon atoms though it is less certain. With this definition of ϵ , the related quantity n^* given by $\epsilon^{1/2}/C$ is the effective principal quantum number. It is obtained from the tables of term values by Bacher and Goudsmit.³¹

The effective quantum numbers are obtained using the standard technique described by White.³² First we proceed to calculate the Rydberg constant R_A for the neon atom; we have

$$R_A = \frac{2\pi^2 m e^4}{h^3 (1+m/AM_p)} \left(\frac{1}{n_1^2} - \frac{1}{n_2^2} \right)$$

where A is atomic weight and M_p is $1/16$ of mass of oxygen atom. Hence, in terms of the Rydberg constant R_∞

$$R_A = \frac{R_\infty}{1+m/AM_p}$$

From the value of R_{∞} by Birge,³³ the most probable value of R_{∞} is

$$R_{\infty} = 109737424 \text{ cm}^{-1}.$$

Therefore,

$$R_{20} = 10973442 \text{ cm}^{-1}$$

for neon atoms. The effective quantum number n^* is defined by

$$T = R/n^{*2},$$

where T is the term value of the energy level of interest. Using the term values from the table by Bacher and Goudsmit, we get

$$n_2^* = 1.65 \text{ and } n_3^* = 2.13.$$

From Table II of Bates and Damgaard, the value of

$\mathcal{F}(n^*, l) = \mathcal{F}(2.12, 1)$ is 3.358, and from Table III, the value of $\mathcal{F}(n_2^*, n_3^*, 1) = \mathcal{F}(1.65, 2.12, 1)$ is 0.778. Hence, from the formula

$$\sigma(n_{l-1}^*, l-1; n_l^*, l; C) = \frac{1}{C} \mathcal{F}(n_l^*, l) \mathcal{F}(n_{l-1}^*, n_l^*, l),$$

it follows that

$$\sigma(1.65, 0; 2.12, 1, 1) = 1 \times 3.358 \times 0.778 = 2.69 a_0^2 = 0.75 \times 10^{-16} \text{ cm}^2.$$

Therefore,

$$\left(\int_0^{\infty} R_i R_j r dr \right)^2 = 3 \times \sigma = 2.25 \times 10^{-16} \text{ cm}^2.$$

III. The Total Cross-Section Calculation

The value of the number of ground-state atoms at room temperature is 1.6×10^{16} atoms/cm³ at a pressure of 0.41 mm of Hg, 2.68×10^{16} atoms/cm³ at a pressure of 0.69 mm of Hg, 4.0×10^{16} atoms/cm³ at a pressure of 1.03 mm of Hg, and 6.5×10^{16} atoms/cm³ at a pressure of 1.68-mm of Hg.

The area of the electron beam is 2.86 cm^2 . Therefore, the flux of the electron beam is given by (i/A_e) . At the pressure of 0.41 mm of Hg, the electron current we used was 1.9×10^{-3} A. Therefore, we get for the flux F of the electron beam, 4.15×10^{15} electrons/cm².

The lifetime τ is obtained from the half-life by the relation

$\tau = \tau_{1/2}/0.693$. For $p = 0.41$ mm of Hg, $\tau = 0.62$ msec. Therefore, we get $1/N_0(J/e) \tau = 1/(4.12 \times 10^{28})$. Hence, we have

$$\sigma_t = \frac{2.88 \times 10^{11}}{4.12 \times 10^{28}} \times \left(\frac{\Delta I}{I} \right) = 0.7 \times 10^{-17} \times \left(\frac{\Delta I}{I} \right) \text{ cm}^2.$$

Similarly, for the pressure of 0.69 mm of Hg, the current we used was 2.2 mA. The lifetime obtained from the half-life is 0.652 msec. Therefore, we have $\sigma_t = 0.346 \times 10^{-17} (\Delta I/I) \text{ cm}^2$.

For $p = 1.03$ mm of Hg, we get $i = 1.9$ mA and $\tau = 0.72$ msec. Therefore, we have

$$\sigma_t = 0.24 \times 10^{-17} \left(\frac{\Delta I}{I} \right) \text{ cm}^2.$$

For $p = 1.68$ mm of Hg, we get $i = 2.15$ mA and $\tau = 0.65$ msec. Therefore, we have

$$\sigma_t = 0.15 \times 10^{-17} \left(\frac{\Delta I}{I} \right) \text{ cm}^2.$$

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