have $L=n\lambda$, where *n* is the mean number of collisions an atom makes before escape, and λ is the mean distance between collisions. It can be shown that $n = (A_b + A_a)/A_a \simeq A_b/A_a$. Equating these two expressions for L then yields

$$\lambda = 4V_b/A_b. \tag{30}$$

Application of this formula to a few simple geometries gives:

Sphere of radius $R: \lambda = 4R/3$.

Cylinder of radius R and length $L: \lambda = 2R/(1+R/2L)$.

Cube of side $L: \lambda = 2L/3$.

PHYSICAL REVIEW

VOLUME 138, NUMBER 4A

17 MAY 1965

Radio-Frequency Resonance of the Metastable State $(2^2 P_{3/2} \ 3^2 S_{1/2})_2$ of Neon Produced and Aligned by Electron Impact*

TETSUO HADEISHI, ORILLA A. MCHARRIS, AND WILLIAM A. NIERENBERG Lawrence Radiation Laboratory, University of California, Berkeley, California (Received 28 December 1964)

The metastable state $(2^2P_{3/2} 3^2S_{1/2})_2$ of neon was produced and aligned by electron impact. The diodestructure electron gun was operated at a high electron-current density under space-charge neutralization conditions. Radio-frequency resonance of the aligned metastable state $(2^2P_{3/2} 3^2S_{1/2})_2$ was observed by monitoring the change of absorption of the λ 6143-resonance radiation passing through the gap between the cathode and anode. The neon metastable-state resonance was compared with the mercury metastablestate $6^{3}P_{2}$ radio-frequency resonance obtained by using the same gun for both, and observed by monitoring the absorption of the λ 5461-resonance radiation passing through the same excitation region. The observed $(g_J)_{Ne}/(g_J)_{Hg}$ was 1.008±0.024. This method could be applied to other atoms having a metastable state with J > 0.

I. INTRODUCTION

LTHOUGH the lifetime of the metastable state A of noble gases such as He, Ne, and Ar is known to be a few milliseconds under a gas pressure of about a millimeter of Hg,^{1,2} no optical detection of rf resonance other than that for helium has been reported up to the present.

One of the major difficulties in determining the rf resonance of the metastable state of the noble gases other than helium by the optical-pumping method is that their parent state has a P configuration. A state having a P configuration has a very fast relaxation time of the alignment at high buffer-gas pressure of about 1 mm Hg, even though the buffer gas is in the ${}^{1}S_{0}$ ground state.³ This requires a very fast optical-pumping speed.

The use of a collimating tube at the bulb entrance will increase the lifetime as described by Eq. (31) in

the text. It also modifies λ , since the derivation above

assumes a constant density throughout the bulb. In the

neck, the density falls uniformly from the equilibrium

density to 0, so that the average density is one-half that in the bulb. Since the rate of wall collision is proportional to the density, the mean wall collision rate is increased by a factor $1+\frac{1}{2}(V_t/V_b)$, where V_t is equal to

the volume of the collimating tube and we have assumed

 $V_i \ll V_b$. The distance between collisions is then given by

 $\lambda = \frac{4V_b}{A_b} \frac{1}{1 + \frac{1}{2}(V_t/V_b)}.$

We recently reported the optical detection of rf resonance of the neon metastable state $(2^2 P_{3/2} \ 3^2 S_{1/2})_2$ by means of production and alignment by electron impact.⁴ The method of producing and aligning the metastable state by high current-density electrons in fact removes the difficulties associated with the requirement for fast optical-pumping speed and a high buffergas pressure of about 1 mm Hg, since the neon metastable state is aligned during the electron transit time corresponding to about the atomic diameter. This report presents the details of the experiment.

II. METHOD AND APPARATUS

The neon atoms were excited to the metastable state from the ${}^{1}S_{0}$ ground state by electron impact. The alignment due to the electron impact may be interpreted

^{*} This work was done under the auspices of the U. S. Atomic Energy Commission.

¹A. V. Phelps and J. P. Molnar, Phys. Rev. **89**, 1202 (1953), and A. V. Phelps and J. L. Pack, Rev. Mod. Phys. **26**, 45 (1955). ² Tetsuo Hadeishi, Ph.D. thesis, Lawrence Radiation Labora-tory Report UCRL-10477, 1962 (unpublished), and T. Hadeishi, S. S. Alpert, and W. A. Nierenberg, Bull. Am. Phys. Soc. **8**, 363 (1962) (1963).

⁴ Peter L. Bender, Joint Institute for Laboratory Astrophysics, University of Colorado (private communication). ⁴ Tetsuo Hadeishi, Orilla A. McHarris, and William A. Nieren-berg, Bull. Am. Phys. Soc. 9, 625 (1964).



approximately according to the explanation made by Bethe⁵ and Oppenheimer.⁶ The probability of the excitation of the atomic level indicated by n is given by

$$F(\mathbf{p}-\mathbf{p}') = -\int u_n^* u_0 \sum_j e^{(i/\hbar)(\mathbf{p}-\mathbf{p}')\cdot\mathbf{r}_j} dr$$

where \mathbf{p} and \mathbf{p}' are the linear momentum of the electron before and after collision with the atom. For electron energies near the threshold of excitation, \mathbf{p}' is zero; therefore, $\mathbf{p} - \mathbf{p}'$ is in the direction of \mathbf{p} . This corresponds to the case in which $\Delta M_j = 0$. In the case of neon, the ground state is ${}^{1}S_{0}$. Therefore, the $M_{J}=0$ of the metastable state will be produced more preferentially if one neglects the electron-exchange effect. Electron exchange results in $\Delta M_J = \pm 1$.

The alignment property of the metastable state can be detected by means of absorption of resonance radiation corresponding to the transition from the metastable state to the higher state. Consideration of spectral line separation, intensity, and transition probabilities leads us to choose the $\lambda 6143$ line.

The polarization ratio is defined as

$$P = (\Delta I_{1} - \Delta I_{11}) / (\Delta I_{11} + \Delta I_{1})$$

where ΔI_{11} and ΔI_{1} are the absorbed intensity of the resonance radiation whose electric vector is, respectively, parallel to and perpendicular to the direction



FIG. 2. Relevant energy levels used in the experiment.

⁵ H. Bethe, Handbuch der Physik, edited by S. Flügge (Springer-Verlag, Berlin, 1933), Vol. 24/1, p. 508. ⁶ J. R. Oppenheimer, Z. Physik 43, 27 (1927).

of the electron beam. This ratio can be expressed in terms of population of a magnetic sublevel as

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

where N_2 , N_1 , and N_0 are the populations of $M_J = \pm 2$, ± 1 , and 0. Therefore, preferential population of the magnetic sublevels of $M_J = 0$ and ± 1 causes P to exceed zero. An rf resonance with sufficiently high H_{rf} equalizes the population distribution, which amounts to causing P to equal zero. Figure 1 shows the relative directions of the lights and the electron beam.

Since the transition probability for $\Delta M_J = 0$ corresponding to the electric vector of resonance radiation parallel to the direction of the electron beam is 4:1:0 for $M_J = \pm 2$ to $\pm 2, \pm 1$ to ± 1 and 0 to 0, the absorption of the resonance radiation at rf resonance increases (Fig. 2).

A. Production and Alignment of the **Metastable State**

To observe the rf resonance, one must obtain a reasonable amount of the metastable-state atoms, and the metastable state must be aligned. The metastable state density was estimated by first measuring the excitation cross section at a pressure of about 1 mm of Hg. Since the rate equation for the metastable state Ncan be expressed by $dN/dt = \sigma(J/e)N_0 - (1/\tau)N$, where J/e is the ratio of electron current density to the electronic charge corresponding to the flux of the electron beam, τ is the relaxation time of the metastable state, N_0 is the ground-state density, and σ is the total excitation cross section. The cross section can be obtained through the relation $\sigma = (N/\tau)/(J/e)N_0$ at equilibrium. The relaxation time τ is measured from the slope of the absorption curve as we turn the electron beam off and on.² The density N of the metastable state is measured from the ratio of the absorbed-light intensity to the incident-light intensity through the relation r

$$\frac{\Delta I}{I} = \frac{l \int k_{\nu} d\nu}{\Delta \nu} = \left(\frac{l}{\Delta \nu}\right) \frac{\pi e^2 N}{m_0 c} f_{ij},$$

where Δv is the half-width of the lamp, t is the absorption path length, and f_{ij} is the oscillator strength. Therefore, the excitation cross section is

$$\sigma = \frac{1}{(J/e)N_0} \times \left(\frac{\Delta I}{I}\right) \times \frac{1}{\tau} \times \frac{1}{(l/\Delta\nu)(\pi e^2/m_0 c)f_{ij}}$$

Using measured values of $\Delta \nu$, $\Delta I/I$, and τ and a value of $f_{ii}=0.22$ calculated by using a method of Bates and Damgaard,⁷ we get $\sigma \sim 10^{-19}$ cm². Therefore,

⁷ D. R. Bates and A. Damgaard, Phil. Trans. Roy. Soc. A242, 101 (1949).

in order to get a metastable-state density of about 8×10^9 atoms/cc corresponding to the pressure of about 10^{-7} mm of Hg at the ground-state gas pressure of about 10^{-2} mm of Hg, we need a current density $J = Ne/N_0\sigma\tau$ of about 16 mA/cm².

For the $\frac{3}{4}$ -in.-diam cathode we used, we needed a current of at least 30 mA to produce the metastablestate density corresponding to 10^{-7} mm of Hg. This pressure was chosen because this operating density is typical for alkali-atom optical-pumping experiments.

Using the space-charge-neutralization method,⁸ we obtained at least 30 mA of electron current over the interaction region about 2 cm along the electron path at the electron energy near the threshold of excitation. In order to operate the high-current electron beam near the 16.5-V threshold of excitation, mercury ions were chosen to space-charge-neutralize the electrons, since the ionization potential of mercury is only 10.4 V. Thus we could change the energy of the electron from 15 to 30 V with electron currents up to 1 A for a $\frac{3}{4}$ -in. cathode, although normally we operated at a much lower current. Under such a circumstance, the cathode is surrounded by a mercury-ion sheath, and the rest of the volume is filled by nearly field-free plasma. The voltage difference between the cathode and anode is concentrated between the cathode and ion sheath, so that the electrons receive most of their acceleration between the cathode and sheath, and enter the nearly field-free plasma perpendicular to the cathode surface, except at the edge of the cathode. The edge effect is minimized by an aperture stop. The electron path can be seen from the mercury radiation emitted from the



Fig. 3. Cross-sectional view of electron-gun excitation tube. A Ne gas pressure of about 1.2×10^{-2} mm is used for the experiment.

plasma region. Depending on the design of the electron gun, either convergence or divergence of the electron beam is observed. By trial-and-error changing the cathode-to-anode gap and placing different aperture stops at various locations, we were able to make the beam trajectory reasonably parallel. Figure 3 shows a cross section of the electron-gun excitation tube.

The parallelness of the electron beam was first tested by resonating the $6^{3}P_{2}$ state of a mercury atom by monitoring the change of the absorption of the λ 5461-



FIG. 4. Block diagram of the detection system.

⁸ I. Langmuir, Phys. Rev. 33, 954 (1929).



FIG. 5. Typical neon metastable-state resonance when the electric vector of $\lambda 6143$ resonance radiation is parallel to the direction of the electron beam.

resonance radiation corresponding to a 6^3P_2 to 7^3S_1 transition.⁹ This resonance was also used to compare the resonance of the neon metastable state, since with the heater current running at 7 A, it is quite difficult to determine accurately the magnetic field at the metastable-state production region of the electron gun. After observing the mercury resonance, neon gas at about 1.2×10^{-2} mm of Hg was admitted into the tube. The neon metastable-state resonance was observed by monitoring the change of the absorption of $\lambda 6143$ -resonance radiation passing though the same region as that of the mercury resonance. A high-vacuum system using all bakeable components was employed.

B. Detection System

The signal-to-noise ratio (S/N) of the neon metastable-state resonance was quite small, although that for mercury was quite large. This necessitated long-time averaging by assigning a field value to the channels of the multichannel pulse-height analyzer and accumulating the signals, which were converted to pulses of varying height corresponding to the channel number. Typically, an integration time of about $2\frac{1}{2}$ h was required for the neon metastable state. The S/N of mercury was high enough so that one could observe it directly from the photomultiplier output in the cathoderay oscilloscope. However, this output was also fed into the pulse-height analyzer in order to compare the resonance points and to calibrate the magnetic-field sweep region. The block diagram is shown in Fig. 4.

The light source used is the usual electrodeless discharge by means of a microwave cavity operating at about 60 to 100 W at 2.45 kMc. Other types of radio-frequency discharge with power up to 500 W were used. However, the microwave discharge turned out to be most convenient because of compactness of the cavity, although any other means of discharge would probably work as well. The required $H_{\rm rf}$ was about 100



FIG. 6. Typical mercury metastable-state resonance when the electric vector of λ 5461 resonance radiation is parallel to the direction of the electron beam.

to 250 mG. Some care was taken to reduce the reactance of the radio-frequency loop in order to avoid the influence of the electric field on the plasma.

III. RESULT

Figure 5 shows the typical neon metastable-state resonance when the polarization is parallel to the direction of the electron beam at a data-accumulation time of about $2\frac{1}{2}$ h and a neon pressure of 1.2×10^{-2} mm of Hg.

This resonance corresponds to an increase in absorption at the rf resonance. In the case of the mercury resonance using the resonance absorption of λ 5461, the absorption of the resonance radiation decreases at the rf resonance when the light is polarized parallel to the electron beam, since the absorption is from 6^3P_2 to 7^3S_1 , i.e., J changes from 2 to 1. This effect is seen from the reversal of the slope in Fig. 6.

The observed value for $(g_J)_{Ne}/(g_J)_{Hg}$ is 1.008±0.024. We believe that this technique demonstrates the possibility of rf resonance of other noble-gas atoms. Further improvement in electron trajectory at much lower gas pressure as well as miniaturization of the electron-gun excitation tube is in progress in order to observe the hyperfine structure of Ar³⁹ by using this technique.

ACKNOWLEDGMENTS

The authors express their appreciation for the help of Professor Edgar Lipworth at the beginning stage of this experiment. Also, he originally suggested this experiment. We also appreciate the help of Dr. S. S. Alpert and Dr. M. B. White who participated in this experiment at the early stage. Michiyuki Nakamura of the Nuclear Chemistry Instrumentation Department designed the electronic detection system, Wilton Berlund of the glass shop constructed the most important part of this experiment—the electron guns, and Douglas Macdonald designed the high-vacuum system.

⁹ H. G. Dehmelt, Phys. Rev. 103, 1125 (1956).