

FIG. 1. Production of electrons in helium.

duction of electrons by metastables becomes negligible, and the electron density decays according to the first term of Eq. (3). We extrapolate this terminal slope toward zero time and take the difference between the actual curve and the extrapolated curve. This yields the second term of Eq. (3), which is shown plotted in Fig. 2.

If we consider the volume loss of metastables to arise from binary collisions between metastables and normal atoms, we may

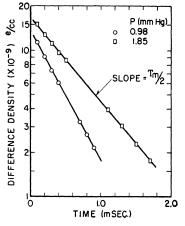


FIG. 2. Loss of metastables in helium.

replace the term  $\nu_d$  of Eq. (2) by Cp, where C is a constant, and p is the gas pressure.<sup>3</sup> Multiplying Eq. (2) by p we have

$$p/T_m = (D_m p/\Lambda^2) + C p^2.$$
<sup>(5)</sup>

In Fig. 3 are plotted values of  $p/T_m$  vs  $p^2$ . The intercept of the curve gives the value  $D_m p/\Lambda^2$ , and the slope of the curve yields C. The diffusion takes place in a container with diffusion length  $\Lambda = 0.735 \text{ cm}^2$  From Fig. 3 we find  $D_m p = 520 \pm 20 \text{ (cm}^2/\text{sec)} \cdot (\text{mm})$ Hg) and  $C = 55 \pm 6$  (mm Hg·sec)<sup>-1</sup>. Runs on several samples of helium give values differing from these results by less than ten percent. Optical absorption experiments by Ebbinghaus yield values of  $D_m p = 535$  and C = 107.4 The difference in the values of the volume loss coefficient, C, may be attributed to differences in gas purity. With specially purified helium<sup>5</sup> (estimated impurity <1:10<sup>8</sup>), we obtain the value C = 55, given above; however, with Airco reagent helium (estimated impurity  $\sim 1:10^4$ ), we obtained

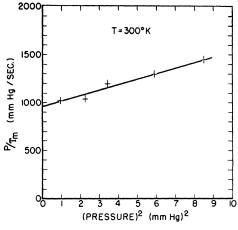


FIG. 3. Diffusion and volume loss of metastables in helium.

a value C = 100. A more complete investigation of the volume loss of metastables is in progress.

Data similar to that shown in Figs. 1-3 have been obtained for neon, giving values of  $D_m p = 200 \pm 20$  and  $C = 230 \pm 20$ . These results may be compared with Phelps' and Molnar's<sup>6</sup> value of  $D_m p = 150 \pm 30$ , obtained by optical absorption. The rather good agreement between the diffusion coefficients obtained from the present measurements and from the optical absorption experiments indicates that the proposed metastable-metastable origin of the ionization is correct. Independent confirmation of the metastable origin of the ionization has been obtained by irradiating the discharge with light of the proper wavelength to raise a metastable atom to a radiating state. A decrease in the metastable concentration results, and we observe a corresponding decrease in the electron density rise of Fig. 1.

In a recent article Johnson et al.7 reported on electron density measurements in helium. They explain their failure to observe an increase in density following the discharge by the fact that their higher electron densities and gas pressures destroy the metastables. Actually, the 10-µsec discharge period they use is too short to permit the metastables to reach more than a fraction of a percent of their equilibrium concentration. As we decrease our discharge time from its usual value of 300  $\mu$ sec to 10  $\mu$ sec, we find that the initial density rise diminishes and finally disappears.

The author wishes to thank Dr. T. Holstein for his assistance during the course of this work

<sup>1</sup> M. A. Biondi, "Measurement of the electron density in ionized gases by microwave techniques," Rev. Sci. Instr., to be published. The apparatus is similar in principle to that described by M. A. Biondi and S. C. Brown, Phys. Rev. **75**, 1700 (1949). <sup>2</sup> M. A. Biondi, Phys. Rev. **79**, 733 (1950), and the references cited theoriem

<sup>4</sup> M. A. Bionin, Phys. Lett.
 <sup>5</sup> See Mitchell and Zemansky, Resonance Radiation and Excited Atoms (Cambridge University Press, London, 1934), pp. 246 ff.
 <sup>6</sup> Ebbinghaus, Ann. Physik 7, 267 (1930); and reference 3, p. 249.
 <sup>5</sup> M. A. Biondi, "Preparation of extremely pure helium gas," Rev. Sci.

Instr., to be published. <sup>6</sup>A. V. Phelps and J. P. Molnar, Bell Telephone Laboratories, private

communication. <sup>7</sup> Johnson, McClure, and Holt, Phys. Rev. 80, 376 (1950).

## The Spin of O<sup>18</sup>

S. L. MILLER,\* A. JAVAN, AND C. H. TOWNES Columbia Radiation Laboratory, Columbia University, New York, New York† (Received March 12, 1951)

BSERVATION of the microwave absorption spectrum of O<sup>16</sup>O<sup>18</sup> and O<sup>18</sup>O<sup>18</sup> in the 5-mm region shows that the spin of O18 is zero.

In the region between 58,300 Mc and 59,900 Mc, all of the expected O<sup>16</sup>O<sup>18</sup> and O<sup>18</sup>O<sup>18</sup> lines have been observed and identified.

TABLE I. Oxygen lines between 58,300 Mc and 59,900 Mc.

Molecule	Transition	Frequency (Mc		
O16O18	$K = 9$ $J = 8 \rightarrow 9$	58.310		
Q16Q16	$K = 9$ $J = 8 \rightarrow 9$	58,324		
Q16Q16	$K = 3$ $J = 4 \rightarrow 3$	58.446		
O16O18	$K = 3$ $J = 4 \rightarrow 3$	58.650		
Q16Q18	$K = 8$ $J = 7 \rightarrow 8$	58,670		
Q18Q18	$K = 3$ $J = 4 \rightarrow 3$	58,900		
O18O18	$K = 7$ $J = 6 \rightarrow 7$	58,965		
O16O18	$K = 7$ $J = 6 \rightarrow 7$	59.075		
O16O16	$K = 7$ $J = 6 \rightarrow 7$	59,163		
O16O18	$K = 4$ $J = 5 \rightarrow 4$	59,220		
Q16Q18	$K = 6$ $J = 5 \rightarrow 6$	59,540		
O16O16	$K = 5$ $J = 6 \rightarrow 5$	59,590		
O16O18	$K = 5$ $J = 6 \rightarrow 5$	59,685		
O18O18	$K = 5$ $J = 6 \rightarrow 5$	59,810		
Q18Q18	$K = 5$ $J = 4 \rightarrow 5$	59,875		

These are listed in Table I. Their positions are predicted closely by Schlapp's formula, as modified by Burkhalter et al.<sup>1</sup> with appropriate changes in constants because of the change in isotopic masses.

A sample of enriched oxygen was very kindly supplied by Professor A. O. Nier. After a brief discharge was run in this gas to approximate an equilibrium distribution, the relative intensities of O<sup>18</sup>O<sup>18</sup> to O<sup>16</sup>O<sup>18</sup> to O<sup>16</sup>O<sup>16</sup> lines were roughly 1:5:15, as may be

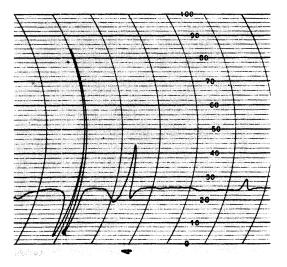


FIG. 1. The K = 7,  $J = 6 \rightarrow 7$  transition for the isotopic combinations O<sup>16</sup>O<sup>16</sup>, O<sup>16</sup>O<sup>18</sup>, and O<sup>18</sup>O<sup>18</sup> (from left to right).

seen from Fig. 1. The three lines shown are the K=7,  $J=6\rightarrow7$ transition for the three different isotopic combinations.

Other observations<sup>2</sup> on O<sup>18</sup> had failed to disclose quadrupole hyperfine structure, and the O<sup>16</sup>O<sup>18</sup> lines show no evidence of magnetic hyperfine structure, which indicates a zero spin for O<sup>18</sup>. However, real proof that the O18 spin is zero must rest on the method of alternating intensities in the homonuclear molecule O18O18

The K=8,  $J=7\rightarrow 8$  transition, which should be at about 58,600 Mc; the K=6,  $J=5\rightarrow 6$  transition at about 59,375 Mc; and the K=4,  $J=5\rightarrow 4$  transition at about 59,400 Mc for O<sup>18</sup>O<sup>18</sup> are all missing. Figure 2 shows the region of the latter two missing lines, bounded by the corresponding transitions for O<sup>16</sup>O<sup>18</sup> which are off scale. In the case of spin=1, these missing lines should be approximately 15 units high, and for higher spin values still stronger. Therefore, it can be concluded that the spin of O<sup>18</sup> is zero, since the alternate rotational levels of O18O18 do not exist. The O18 nucleus also follows Bose-Einstein statistics, as does O16, since the same rotational levels are forbidden in both cases.

Magnetic hyperfine structure has been found in the same sample

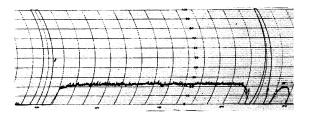


FIG. 2. Recording trace from the K=4,  $J=5\rightarrow 4$  transition for  $O^{18}O^{18}$ (on the right) to the K=6,  $J=5\rightarrow 6$  transition for  $O^{18}O^{18}$  (on the left). The missing  $O^{18}O^{18}$  lines K=4,  $J=5\rightarrow 4$  and K=6,  $J=5\rightarrow 6$  should be about midway between them.

for O<sup>16</sup>O<sup>17</sup>. Although no definite spin determination has as yet been made from this structure, the O17 spin is evidently greater than 1/2.

The microwave lines were detected with a recording Zeeman modulation spectrometer constructed by A. L. Schawlow, T. M. Sanders, and W. A. Hardy.

AEC Predoctoral Fellow. Work supported partly by the AEC and partly by a joint Signal Corps-<sup>1</sup> Burkhalter, Anderson, Smith, and Gordy, Phys. Rev. 79, 651 (1950).
 <sup>2</sup> Townes, Holden, and Merritt, Phys. Rev. 74, 1113 (1948).

## Evidence for the Formation of Positronium in Gases\*

MARTIN DEUTSCH

Laboratory for Nuclear Science and Engineering, and Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts (Received March 13, 1951)

<sup>¶</sup>HE distribution of time delays between the emission of a nuclear gamma-ray from the decay of Na<sup>22</sup> and the appearance of an annihilation quantum has been measured for positrons stopping in a large number of gases and gas mixtures, extending earlier measurements.1 A complete interpretation of the results appears to be fairly complex and will be attempted in a more extensive communication. At this time we want to report on some definite proof of the abundant formation of positronium, the bound electron-positron system analogous to the hydrogen atom. The ground state of this atom is expected<sup>2</sup> to have a lifetime against two-quantum annihilation of about 10<sup>-10</sup> sec if the spins are antiparallel and slightly over  $10^{-7}$  sec if they are parallel,<sup>3</sup> decaying by three-quantum annihilation. Ore4 has shown that, in general, there is no mechanism for the rapid destruction of the triplet state at low energies. One may expect, however, that in a gas containing molecules with an odd number of electrons, such as nitric oxide, the triplet state would be converted very rapidly to the singlet by an electron exchange. In NO this requires essentially no energy at room temperature, the energy difference for the two spin orientation being only about 13 millivolts. The cross section for this process may be very large, since it is a resonance effect involving the coulomb interaction rather than a magnetic conversion. (The latter is expected to be very slow.<sup>4</sup>) Thus, even a small admixture of NO should cause the very rapid annihilation of those positrons which would otherwise have decayed by three-quantum annihilation with a period of the order of  $10^{-7}$  sec. Figure 1 shows this phenomenon. In nitrogen the number of delayed counts is reduced by a factor of three by the addition of 3 percent NO. These counts then appear instead in the "prompt" channel, not shown in Fig. 1. In freon the effect is even greater, hardly any delayed coincidences remaining upon addition of NO. The residue in  $N_2$  is probably due to free positrons.

Oxygen has a similar but less pronounced action. In pure O<sub>2</sub> the number of delayed coincidences is about half as great as in N2 (Fig. 1).

Confirmation of the above interpretation comes from the direct observation of the continuous gamma-ray spectrum due to the

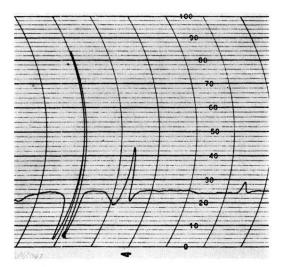


FIG. 1. The K = 7,  $J = 6 \rightarrow 7$  transition for the isotopic combinations O<sup>16</sup>O<sup>16</sup>, O<sup>18</sup>O<sup>18</sup>, and O<sup>18</sup>O<sup>18</sup> (from left to right).

11	11 1	1	1	-1-		- 1	1	-1			1	i.		- 11	$\left( \begin{array}{c} i \\ i \end{array} \right)$
	11		1	1			1		1. di	1.	1.11.11				1 1
	11_	ļ				1000	A 1. 28. 297	10.000	1.00	1.40	1	1	Concerne and	Second Second	
	Ľ.							1	1	1			-		
11	11-1			/			I,	I	1.	and the second	I		1		11
+1	11 6	-and	mage	denti	-	فيعيله	min				many	-		-#	1 12

FIG. 2. Recording trace from the K=4,  $J=5\rightarrow 4$  transition for O<sup>16</sup>O<sup>18</sup> (on the right) to the K=6,  $J=5\rightarrow 6$  transition for O<sup>16</sup>O<sup>18</sup> (on the left). The missing O<sup>18</sup>O<sup>18</sup> lines K=4,  $J=5\rightarrow 4$  and K=6,  $J=5\rightarrow 6$  should be about midway between them.