

## Lifetime of the $2^1P_1$ and $3^1P_1$ States of Atomic Helium

James M. Burger and Allen Lurio

*IBM Watson Laboratory, Columbia University, New York, New York 10025*

(Received 17 July 1970)

The radiative lifetime of the  $2^1P_1$  and  $3^1P_1$  state of atomic helium has been measured by the level-crossing technique. In the experiment, a beam of  $2^1S_0$  metastable atoms was first produced and then excited to the  $2^1P_1$  or  $3^1P_1$  state with resonance radiation. The lifetimes were measured by observing the zero-field level-crossing signals in the detected 584- or 537-Å fluorescent radiation when these  $^1P_1$  states decayed to the  $1^1S_0$  ground state. The results are  $\tau(2^1P_1) = (5.57 \pm 0.15) \times 10^{-10}$  sec and  $\tau(3^1P_1) = (1.73 \pm 0.11) \times 10^{-9}$  sec. These values are in excellent agreement with theory.

### I. INTRODUCTION

In recent years, very accurate wave functions for the low-lying levels of the helium atom have been obtained.<sup>1</sup> These wave functions are in excellent agreement with experiment in that they predict the energy level scheme of the observed spectrum to within the experimental accuracy (one part in  $10^7$ ). However, a number of other properties of the helium atom, in particular the lifetimes, which also have been calculated very accurately with these wave functions,<sup>2</sup> have not been experimentally studied in such detail.

In the present work, a new<sup>3</sup> level-crossing technique has been independently developed and used to measure the lifetime of the  $2^1P_1$  and  $3^1P_1$  states of the neutral helium atom. In most previous optical level-crossing work, the experimental procedure has been to excite an initial atomic level with resonance radiation and to observe the magnetic (or electric) field dependence of the fluorescent radiation to this same initial level. However, such a procedure in the case of the  $2^1P_1$  and  $3^1P_1$  levels of helium would require strong stable sources of 584- and 537-Å radiation, respectively, which are not readily available.

To circumvent the need for such vacuum ultraviolet sources, an alternate procedure was devised which made use of the metastability of the  $2^1S_0$  state of helium. An atomic beam of  $2^1S_0$  metastable atoms was first produced by electron bombardment (Fig. 1). The  $2^1S_0$  beam then passed through a uniform variable magnetic field where the  $2^1P_1$  or  $3^1P_1$  state was excited by 20 582-Å ( $2^1S_0-2^1P_1$ ) or 5016-Å ( $2^1S_0-3^1P_1$ ) resonance radiation. The 584- or 537-Å photons, resulting from the dominant decay of the  $2^1P_1$  or  $3^1P_1$  state to the  $1^1S_0$  ground state, were detected. The magnetic field dependence of the level-crossing signal, observed in the detected 584- or 537-Å fluorescent radiation, was used to determine the lifetime via the Breit-Franken formula.

### II. APPARATUS

#### A. General Considerations

The general design of the atomic-beam apparatus (Figs. 2 and 3) followed from several requirements. Since the 584- or 537-Å photons are transmitted by no structural material (and poorly by thin films) and are appreciably absorbed by ground-state  $1^1S_0$  helium atoms at fairly low pressures, the experiment had to be done with an atomic beam in a vacuum of  $10^{-6}$ -mm Hg or better. A two-chamber vacuum system was constructed with a source chamber to produce the metastable beam and an interaction chamber in which the level-crossing experiments were performed. An iron magnet was used, as it both permitted a convenient level-crossing geometry and provided sufficiently intense fields for the scanning of line profiles.<sup>4</sup> With an iron magnet, excitation and detection were necessarily in a plane perpendicular to the magnetic field. Since the windowless electron multiplier for detection of the 584- and 537-Å photons was totally insensitive to the infrared and visible exciting radiation, it was possible for the incident and exiting radiation to be colinear and perpendicular to the metastable atomic beam. For this geometry, the level-crossing signal (Breit-Franken formula) took on a simple form. Also with this arrangement, the straight-through metastable beam could be separately monitored.

#### B. Components

*a. Vacuum system.* The two-chamber vacuum system was constructed of brass, and the source chamber could be slid back for easy access to its interior. A brass separating plate with a 7.9-mm-high by 3.2-mm-wide hole for the beam to emerge was mounted on the source chamber. The chambers were each pumped by a baffled 300 liter/sec oil-diffusion pump; both pumps were backed by a single Welch Model 1402 mechanical pump. Each chamber contained a liquid-nitrogen trap. Source-

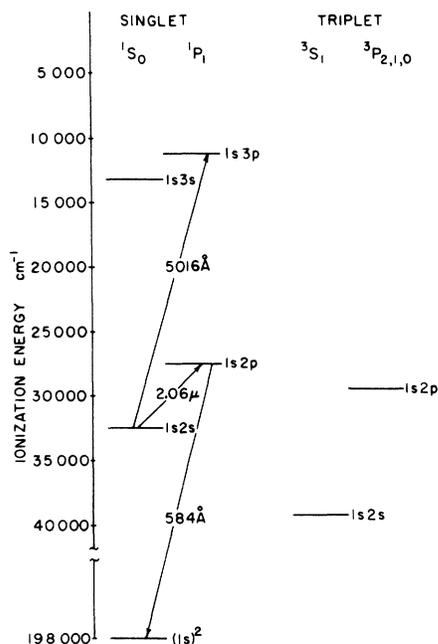


FIG. 1. Low-lying levels of the helium atom.

and-interaction-chamber pressures were read with Veeco RG75P ionization gauges; to read the helium pressure the observed value (nitrogen calibration) was multiplied by 4.8. With liquid-nitrogen trapping (no helium-gas input), typical source-and-interaction chamber pressures were  $10^{-7}$ - and  $3 \times 10^{-7}$ -mm Hg, respectively.

*b. Helium gas.* Helium gas at room temperature was admitted into the source chamber by a Vacronic (East Northport, N. Y.) VV-50 leak valve. The  $\text{He}^4$  gas was of the standard high-purity quality from the Linde Company (Division of Union Carbide) and

was obtained in 1-liter pyrex bulbs at about 1 atm. Under experimental conditions, the helium pressure in the source chamber was  $1.7 \times 10^{-5}$ -mm Hg, corresponding to a flow of  $8 \times 10^{16}$  atoms/sec. The corresponding observed helium partial pressure in the interaction chamber was  $4 \times 10^{-7}$ -mm Hg. The gas flowing into the source chamber was directed into the electron bombarder through a crinkle-foil source to increase the forward intensity over that of the cosine law.<sup>5</sup> Moreover, to compensate for the approximately  $12^\circ$  change in direction of the  $\text{He}^4$  atoms in the inelastic ( $1^1S_0 - 2^1S_0$ ) collisions with electrons,<sup>6</sup> the gas beam was directed into the electron bombarder at an angle of about  $78^\circ$  to the electron velocity.

*c. Electron bombarder.* The electron bombarder, a modification of previous designs,<sup>7</sup> was electrically a triode structure with a cathode, a grid, and a collector (Fig. 4). The entire structure was mounted between the pole pieces of a permanent magnet, which produced a constraining field of 1300 G parallel to the electron current. The type-B cathode (Philips Metalonics, Mt. Vernon, N. Y.) was mounted in a groove on the top of a hollow molybdenum block and had a 1.6-mm-wide by 1.9 cm-long emitting surface. A helical oxide-coated filament was inserted in the cylindrical hole in the cathode structure. The filament was heated by a dc supply and about 40 W (3.2 A) were required for emission.

In operation, the collector-grid voltage was fixed at 5 V and the grid-cathode voltage was varied to obtain a maximum beam. Though not critical, this voltage was typically 45–50 V, consistent with other work.<sup>8</sup> For these conditions, collector currents were of the order of 15 mA, corresponding to a current density of 0.05 A/cm<sup>2</sup>. The estimated<sup>9</sup> fractional  $2^1S_0$  production for an excitation cross section<sup>10</sup> of  $4 \times 10^{-18}$  cm<sup>2</sup> was approximately  $10^{-5}$ .

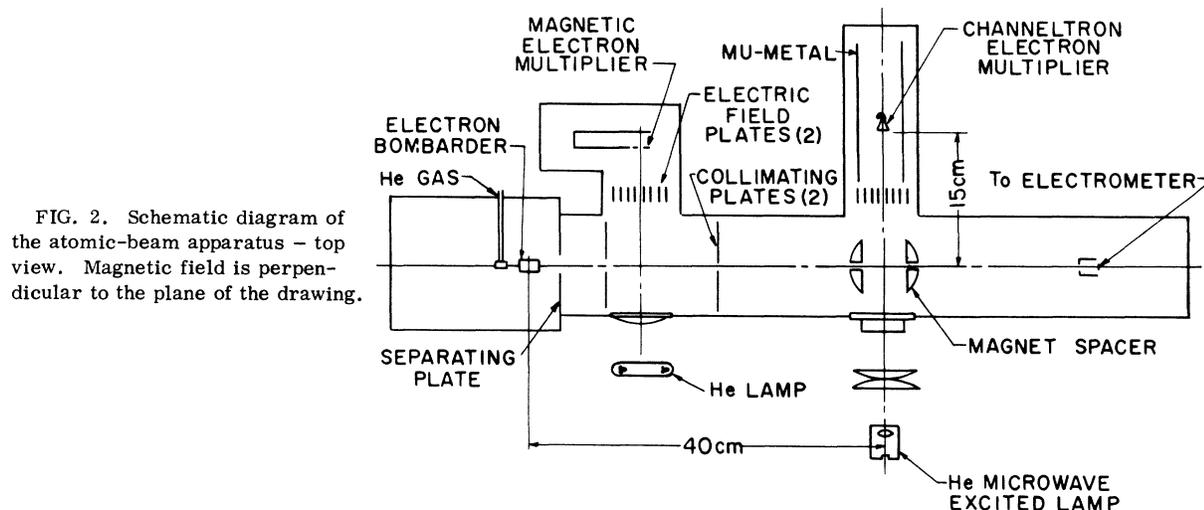


FIG. 2. Schematic diagram of the atomic-beam apparatus - top view. Magnetic field is perpendicular to the plane of the drawing.

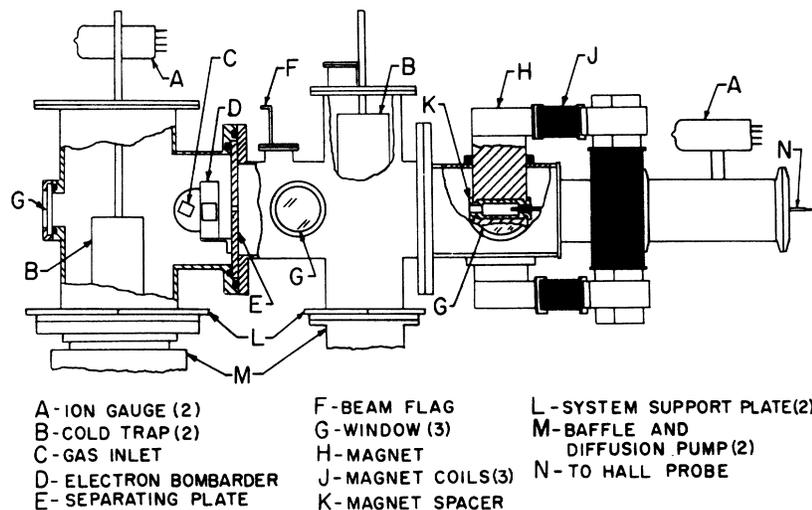


FIG. 3. Structural view of the atomic-beam apparatus - side view.

*d. Beam monitor.* The metastable beam was monitored by measuring the current of ejected electrons which resulted when the metastable atoms impinged on a gold surface. The voltage developed by the collected current across a  $10^{10}\text{-}\Omega$  resistor was read with an electrometer circuit. The use of a gold target permitted an estimate of the metastable beam, since the electron-ejection efficiencies of singlet and triplet metastable helium atoms are known to be approximately one-half and one-quarter, respectively.<sup>11</sup> The gold target is moreover insensitive to  $584\text{-}\text{\AA}$  photons.<sup>11</sup>

*e. Detector.* The recently introduced channeltron electron multiplier (Bendix Corp.) proved to be a most satisfactory detector of the  $584\text{-}\text{\AA}$  and  $537\text{-}\text{\AA}$  photons. Under the operating conditions here (cathode at  $-2850\text{ V}$ ), this detector had a dark current of about 0.1 counts/sec and a gain of  $5 \times 10^7$ . For  $584\text{-}\text{\AA}$  photon detection, the quantum efficiency of the sensitive surface is about 0.2; due to inefficiencies in collecting electrons ejected on the outer portion of the cone, the effective efficiency is about 0.12. The channeltron was mounted by inserting its cone (10 mm) in a hole in a piece of Teflon; two tapered Teflon pieces were pushed against the rear of the cone by screws and held the channeltron firmly in place. The channeltron was magnetically shielded with mu-metal; no magnetic field dependence in the output signal was detected when the channeltron was irradiated by mercury-lamp light to which it is sensitive. The channeltron was also completely shielded from ions by a stack of thin parallel copper plates<sup>12</sup> (3.2 mm apart and 2.5 cm deep) which were alternately biased at 0 and  $+60\text{ V}$ . To minimize the background signal, the channeltron was out of direct line with any wide-angle portion of the beam (Fig. 2).

*f. Magnet.* The magnet (Figs. 2 and 3) was con-

structed of soft iron (Armco) and plated with copper and nickel for protection against rusting. The 7.3-cm-diam pole pieces entered the vacuum system through rubber *O* rings and were held at a fixed separation of 2.5 cm by means of a brass spacer. The atomic beam entered and left the spacer through a 1.6-cm-high by 0.95-cm-wide hole; the orthogonal aperture for incident and exiting radiation was 2.2 cm high by 5 cm long (along the beam). For increased magnetic field homogeneity, each pole piece had a 1.6-mm-high by 3.2-mm-radial-width shim on its outer rim; the field was constant to within 0.3% over the central 3.8-cm portion of the gap. The magnetic field was measured with a transverse Hall probe, specially mounted by F. W. Bell (Columbus, Ohio) at the end of a brass tube; this tube was inserted into a concentric brass tube which extended into the magnet spacer through the exit hole for the atomic beam. The Hall element was located radially 1.9 cm from the magnet gap center.

*g. Lamps.* The helium resonance lamps were microwave-excited quartz bulbs of helium filled to

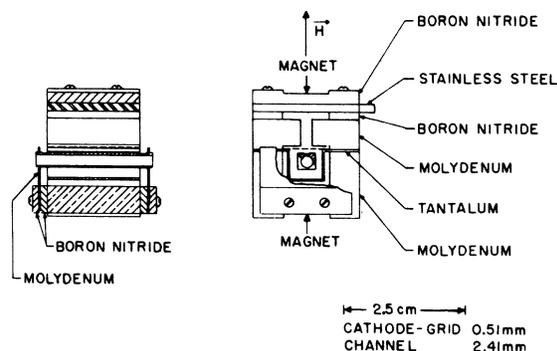


FIG. 4. Electron bombarder.

pressures from 1- to 6-mm Hg. The bulbs, typically 1 cm diam by 2 cm long, were mounted at the end of a microwave cavity by means of a glass stem which protruded through the cavity wall. The tunable Evenson cavity (Ophthos Instr., Rockville, Md.) was driven by a Raytheon Microwave Power Generator PGM10 with a maximum power output of 85 W at 2450 MHz. Typically the lamps were driven at 20% power for 20 582-Å radiation and at about 60% power for 5016-Å radiation; this higher power slightly increased the 5016-Å intensity. These microwave lamps had the advantage of being intense localized sources whose useful intensity was approximately twice that obtained with a standard Osram He<sup>4</sup> lamp.

As subsequently discussed, a black-body source was used to produce 20 582-Å radiation. This source was a Sylvania DXN halogen cycle lamp (sun-gun) with a color temperature of 3400 °K at 1000-W input and was typically driven from a Variac at 85% of line voltage. The 7.6-cm-long bulb was housed in a cylindrical water-cooled jacket and further cooled by a high-velocity stream of air between the jacket and the bulb. The radiation emerged from a 2.2-cm-long by 1-cm-high aperture in the housing.

Due to the sun-gun intensity, unused radiation had to be filtered out. This was accomplished by a germanium filter coated on both faces (2500 Å of silicon nitride) for high transmission in the spectral range of 2 μ. The filter used was opaque below 1.8 μ and had 85% transmission at 2 μ. The germanium itself was a polished disk 3.8 cm diam and 3.2 mm thick; such disks are readily available due to their use as CO<sub>2</sub> laser windows. By use of a low-temperature solder (100-°C melting point) and a special flux, the disk was soldered onto a brass ring through which water for cooling was flowed. The filter was located about 3 cm in front of the surface of the sun-gun bulb and in addition cooled with air on the irradiated surface.

### C. Technique of Measurement

The technique of measurement is indicated in block-diagram form in Fig. 5. A Hewlett-Packard Model 202A Function Generator repetitively supplied a triangular-wave voltage to a Kepco BOP36-5M programmable power supply, which provided the current for the iron magnet. A trigger pulse at the peak of the triangular wave initiated the channel address cycle of a multichannel analyzer, a CAT 1000 (Tech. Meas. Corp., North Haven, Conn.). The CAT completed each cycle slightly before the arrival of the next trigger pulse. Channeltron pulses were counted in one-quarter of the CAT mem-

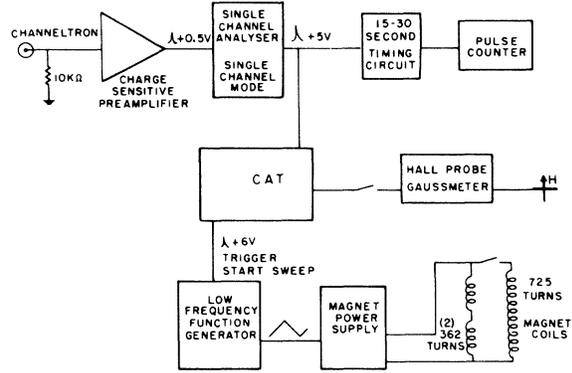


FIG. 5. Block diagram of the signal-averaging technique.

ory (256 addresses); with an address rate of 0.080 sec/channel (to maintain equilibrium in the iron magnet), each sweep took a little over 20 sec. For a period of this duration, drifts in metastable beam and lamp intensity were small and were averaged out over a large number of sweeps. The varying magnetic field was determined by reading the Hall-probe-gaussmeter output into another quarter of the CAT by use of an analog to digital converter; a calibration obtainable from the gaussmeter was read into a third quarter so as to convert counts to magnetic field. One CAT sweep of the gaussmeter output was taken at the beginning and the end of a run to minimize any errors due to slow drifts in magnetic field. The data stored in the CAT was read directly into an IBM 1130 computer in digital form.

The magnetic field was measured with a Bell 640 gaussmeter in combination with a T6010 temperature-compensated probe (F. W. Bell, Columbus, Ohio). This unit had a quoted accuracy in the 0- to 10 000-G range of 0.05% of the full-scale value plus 0.5% of the reading, and was cross-checked against proton resonance (high fields) and a rubidium magnetometer (low fields). The error in field measurement introduced by the use of the CAT was small, as was verified by measuring known static fields.

## III. THEORY

### A. Breit-Franken Formula Evaluation

The Breit-Franken formula gives the spacial and polarization dependence of the rate of scattering of resonance radiation by an atomic system. In general, this formula for the scattered- or decay-photon intensity is<sup>14</sup>

$$R(\theta\phi\alpha; \theta'\phi'\alpha') = C \sum_{mm'\mu\mu'} \frac{\langle am | \hat{\epsilon} \cdot \vec{r} | b\mu' \rangle \langle b\mu' | \hat{\xi} \cdot \vec{r} | cm' \rangle \langle cm' | \hat{\xi} \cdot \vec{r} | b\mu \rangle \langle b\mu | \hat{\epsilon} \cdot \vec{r} | am \rangle}{\Gamma + i(E_{b\mu} - E_{b\mu'})/\hbar}, \quad (1)$$

where  $a, b$ , and  $c$  denote the energy level and angular-momentum quantum numbers for the initial state, the excited state, and the final state, respectively;  $m, \mu$  and  $\mu', m'$  their respective magnetic quantum numbers;  $\Gamma = 1/\tau$ , where  $\tau$  is the lifetime of state  $b$ ;  $\hat{\epsilon}$  and  $\hat{\xi}$  are the incident and exiting polarization vectors, respectively, where the above angles are those of Fig. 6;  $C$  is an unimportant proportionality constant. The magnetic field dependence of the scattered intensity generally permits a lifetime determination of considerable accuracy, provided one has a "white" incident-radiation profile over the region of interest.

In the present experiments with  $\text{He}^4$ , which has zero nuclear spin, the initial state was a  $^1S_0$ , the excited state a  $^1P_1$ , and the final state a (different)  $^1S_0$ . For this case, the above expression as a function of an externally applied magnetic field can be explicitly written in complete generality, since the energy eigenvalues of the  $^1P_1$  magnetic sublevels are  $E_{b\mu} = E_b + g_1\mu_B H\mu$ . This calculation has been described elsewhere.<sup>15</sup> Moreover, the conditions of this experiment yield several simplifications. As the polarization of the 584- or 537-Å fluorescent radiation is not detected, a sum over the final polarization states ( $\alpha'$ ) may be performed to give  $R(\theta\phi\alpha; \theta'\phi')$  in the above notation. Since the channeltron solid angle is small, the emitted photon direction is well defined with the coordinates  $\theta' = \frac{1}{2}\pi, \phi' = \pi$  (Figs. 2 and 6). To account for the angular spread of the incident radiation, this expression must be averaged over solid angle, i.e.,

$$\bar{R}(\theta\phi\alpha; \theta'\phi') = \left( \int_{\theta-\Delta\theta}^{\theta+\Delta\theta} d\phi \int_{\theta-\Delta\theta}^{\theta+\Delta\theta} \sin\theta d\theta \right)^{-1} \times \int_{\theta-\Delta\theta}^{\theta+\Delta\theta} d\phi \int_{\theta-\Delta\theta}^{\theta+\Delta\theta} R(\theta\phi\alpha; \theta'\phi') \sin\theta d\theta. \quad (2)$$

With the assumption of a uniform incident intensity per unit solid angle, the final exact result for the two orthogonal incident polarization directions perpendicular ( $\alpha = \frac{1}{2}\pi$ ) and parallel ( $\alpha = 0$ ) to the magnetic field is

$$\bar{R}\left(\epsilon\phi\frac{\pi}{2}; \frac{\pi}{2}\pi\right) = \frac{f_1}{6\Gamma} \left( 1 + \frac{\sin 2\Delta\phi}{2\Delta\phi} \frac{\cos 2\phi - (2g_1\mu_B H/\hbar\Gamma) \sin 2\phi}{1 + (2g_1\mu_B H/\hbar\Gamma)^2} \right), \quad (3)$$

$$\bar{R}\left(\nu\phi 0; \frac{\pi}{2}\pi\right) = 2 \frac{f_1}{6\Gamma} - \epsilon(\theta, \Delta\theta) \bar{R}\left(\theta\phi\frac{\pi}{2}; \frac{\pi}{2}\pi\right),$$

where

$$\epsilon(\theta, \Delta\theta) = \cos^2\theta \cos^2\Delta\theta + \frac{1}{3} \sin^2\theta \sin^2\Delta\theta$$

and<sup>16</sup>

$$f_1 = C(n_a 0 \| \vec{r} \| n_b 1)(n_b 1 \| \vec{r} \| n_c 0)(n_c 0 \| \vec{r} \| n_b 1)(n_b 1 \| \vec{r} \| n_a 0).$$

For the experimental geometry,  $\theta = \pi/2, \phi \approx 0$ ,

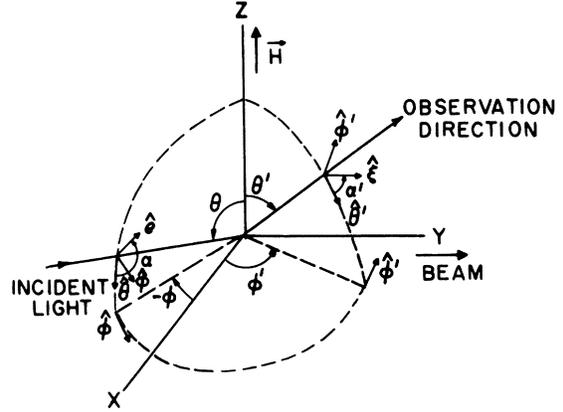


FIG. 6. Coordinate and polarization system used in the quantum-mechanical calculation of the zero-field level-crossing signal. Primed ( $\xi$ ) and unprimed ( $\epsilon$ ) coordinates refer to the exiting and incident radiation, respectively.  $\alpha$  (or  $\alpha'$ ) is the angle between  $\theta$  (or  $\theta'$ ) and  $\hat{\epsilon}$  (or  $\hat{\xi}$ ); it is swept out in the sense of a right-hand screw rotation about an outward normal to the sphere.

the entire field-dependent signal (except for solid-angle effects) arises from the polarization  $\alpha = \pi/2$  (note  $\sin 2\Delta\phi/2\Delta\phi \approx 0.96, \epsilon(\theta, \Delta\theta) \approx 0.02$  for the optics used). This  $\alpha = \pi/2$  signal is a Lorentzian centered at 0 G (strictly only for  $\phi = 0$ ); its value decreases at high fields (in the limit  $\Delta\phi = 0$ ) to 50% of the peak value at 0 G. The  $\alpha = 0$  signal is nearly constant and is about equal to the  $\alpha = \pi/2$  signal at 0 G. Even with arbitrary intensities of the two incident polarizations, the magnetic field dependence of the level-crossing signal in this geometry can be represented by the functional form

$$R = G_2 \left( 1 + \frac{1 - G_1 G_3 H}{1 + G_1^2 H^2} \right) + G_4, \quad (4)$$

where  $G_1 = 2g_1\mu_B/\hbar\Gamma$ . For the lifetime determination, the observed level-crossing signal was fit as a function of magnetic field to this expression with  $G_1, G_2, G_3$ , and  $G_4$  as parameters. The parameters  $G_2, G_3$ , and  $G_4$  described the signal intensity and optical arrangement and provided a useful check on the consistency of the data. The parameter  $G_3 = \tan 2\phi$  gave the angle by which the incident and exiting radiation were not colinear. An APL/360 least-squares program was used for the data analysis.

#### B. Theoretical Radiative Lifetime

The radiative lifetime  $\tau(A)$  of a state  $A$  which decays to states  $B$  is given in terms of the transition probabilities  $A(A, B)$  for the decay channels by the expression<sup>17</sup>

$$\tau(A) = 1/\Gamma(A) = 1/\sum_{B < A} A(A, B). \quad (5)$$

The transition probabilities are in turn each related to the absorption oscillator strengths  $f(A, B)$  by

$$A(A, B) = f(A, B) \left/ \left( \frac{mc}{8\pi^2 e^2} \frac{2J_A + 1}{2J_B + 1} \lambda_{AB}^2 \right) \right., \quad (6)$$

where  $J_A$  and  $J_B$  are the angular momenta,  $\lambda_{AB}$  the vacuum wavelength of the transition,  $c$  the velocity of light, and  $m$  and  $e$  the electronic mass and charge, respectively.

For the cases of the  $2^1P_1$  and  $3^1P_1$  lifetime, the necessary oscillator strengths have been evaluated to a high quoted precision.<sup>2</sup> For the  $2^1P_1$  state,

$$\Gamma(2^1P_1) = A(2^1P_1-1^1S_0) + A(2^1P_1-2^1S_0),$$

with a resulting radiative lifetime

$$\tau(2^1P_1) = 5.555 \times 10^{-10} \text{ sec.}$$

The branching ratio is found to be

$$A(2^1P_1-1^1S_0)/A(2^1P_1-2^1S_0) = 911.$$

For the  $3^1P_1$  lifetime [neglecting the  $3^1S_0$  and  $3^1D_2$  states since  $A(A, B) \propto 1/\lambda_{AB}^2$ ], one has

$$\Gamma(3^1P_1) = A(3^1P_1-1^1S_0) + A(3^1P_1-2^1S_0),$$

with a resulting radiative lifetime

$$\tau(3^1P_1) = 1.726 \times 10^{-9} \text{ sec.}$$

The branching ratio is

$$A(3^1P_1-1^1S_0)/A(3^1P_1-2^1S_0) = 42.3.$$

For the Lorentzian dependence of scattered intensity on magnetic field, a half half-width in gauss is defined by

$$H_{1/2} = \hbar\Gamma/2g_1\mu_B,$$

where

$$g_1 = g_J(1P_1) = 1.00000$$

and  $\mu_B$  is the Bohr magneton. For the above states, we find that

$$H_{1/2}(2^1P_1) = 102.35 \text{ G}, \quad H_{1/2}(3^1P_1) = 32.93 \text{ G}.$$

A half-width in gauss corresponding to the observed lifetime was the quantity directly determined experimentally; in terms of the functional form analyzed by least squares, this half half-width was equal to  $1/G_1$ .

#### C. Effects from Residual Gas

In level-crossing measurements nonradiative contributions to the lifetime also enter the Breit-Franken formula. With an atomic beam, however, increased decay rates due to collisional deexcitation<sup>18</sup> were negligible. Moreover, by operating at pressures such that the absorption of decay photons by ground-state helium atoms was small, differences in the true and observed lifetime

arising from multiple scattering of these decay photons in the magnetic field (coherence narrowing) were nearly eliminated. The modification of the true radiative  $2^1P_1$  lifetime due to coherence narrowing may be estimated,<sup>19</sup> where the small contribution from the  $(2^1P_1-2^1S_0)$  decay is neglected. When coherence narrowing is taken into account, the radiative decay rate  $\Gamma$  in the Breit-Franken formula becomes

$$\Gamma_{\text{obs}} = \Gamma(1 - \frac{7}{10}x),$$

with  $x = 1 - e^{-2KNl}$ . Here  $l$  is the length of the scattering path (taken as 3.8 cm or a little greater than half the magnet-pole diameter);  $N$  is the ground-state helium atom density ( $P = 4 \times 10^{-7}$ -mm Hg), and  $K$  is given by

$$K = (3/32\pi)\lambda^3\Gamma/v = 7.3 \times 10^{-14} \text{ cm}^2,$$

where  $\lambda$  is 584 Å and  $v$ , the atomic velocity, is  $1.5 \times 10^5$  cm/sec. With these numbers, we get

$$\Gamma_{\text{obs}}(2^1P_1) = \Gamma(2^1P_1)(1 - 0.004).$$

For the  $3^1P_1$  lifetime, we find

$$K(3^1P_1) \simeq K(2^1P_1)\Gamma(3^1P_1)/\Gamma(2^1P_1),$$

neglecting the slight wavelength difference, and hence at this pressure, we get

$$\Gamma_{\text{obs}}(3^1P_1) = \Gamma(3^1P_1)(1 - 0.001).$$

The absorption of the decay photons between the excitation region and the channeltron detector, a distance  $L$  of 15 cm, may be estimated for the above pressure. In the case of a Gaussian source and Gaussian absorption profile, the fractional absorption  $\Delta I/I = N\sigma L$ , where<sup>20</sup>

$$\sigma = r_0 c f [2(\pi \ln 2)^{1/2}] / [(\Delta\nu^2 + \Delta\nu_D^2)^{1/2}], \quad (7)$$

with  $r_0 = e^2/mc^2$ ,  $c$  the velocity of light,  $f$  the absorption oscillator strength, and  $\Delta\nu$  and  $\Delta\nu_D$  the full width at half-maximum of the source and the absorber profiles, respectively. The source photons, in this case the decay photons from the beam observed perpendicular to the beam velocity, have a negligible width compared to the residual helium-gas absorption width, so that  $\Delta\nu_D \gg \Delta\nu$ . For a residual helium-gas temperature of 300 °K, we get

$$\Delta\nu_D = (2\nu/c) [(2 \ln 2)KT/M]^{1/2} = 31 \times 10^9 \text{ Hz}.$$

Then with  $f(2^1P_1-1^1S_0)$  from theory,<sup>2</sup> we find

$$\sigma(1^1S_0-2^1P_1) = 2.1 \times 10^{-13} \text{ cm}^2$$

and

$$\Delta I/584/I584 = 0.03.$$

Similarly, the fractional absorption of the 537-Å photons can be estimated as 0.01. Most of this absorption and scattering of decay photons does

not contribute to coherence narrowing since it is in a region of essentially zero magnetic field.

#### D. Effect of Lamp Profile

As previously indicated, the Breit-Franken formula is based on the assumption of an incident white-light profile. However, in the case of ( $2^1P_1-2^1S_0$ ) radiation from a helium resonance lamp, the intensity profile can be seen to show considerable variation over a range of several hundred gauss about the center.<sup>4</sup> As an order-of-magnitude estimate, consider the lamp intensity at the ( $2^1P_1, m_J = \pm 1$ )- $2^1S_0$  resonant frequency when the magnetic sublevels have been Zeeman shifted by 1.5 radiative half half-widths [ $H_{1/2}(2^1P_1) = 102.35$  G]. With the assumption of a Gaussian profile of incident radiation with a full width at half-maximum  $\Delta\nu = 1.7 \times 10^9$  Hz (effective He<sup>4</sup> lamp temperature 850 °C), the ratio of this intensity to that at the lamp center is

$$\exp\left[-\left(\frac{2(\ln 2)^{1/2}(\nu - \nu_0)}{\Delta\nu}\right)^2\right] = 1 - 0.04 \quad (8)$$

Hence, the effect of helium-resonance-lamp profile could be expected to cause an error of several percent in a  $2^1P_1$  lifetime measurement based on data taken over a magnetic field range,  $|H| \leq 154$  G, and analyzed by the Breit-Franken formula. A further discussion of the lamp profile and its effect on the  $2^1P_1$  lifetime has been given elsewhere.<sup>21</sup> For the case of the ( $3^1P_1-2^1S_0$ ) resonance line from a lamp at the same effective temperature ( $\Delta\nu = 7 \times 10^9$  Hz), the intensity at 2 radiative half half-widths [ $H_{1/2}(3^1P_1) = 32.93$  G] has decreased by 0.05% and hence for purposes of a  $3^1P_1$  lifetime determination, the 5016-Å line from a helium resonance lamp can be considered "white."

#### IV. BEAM AND BACKGROUND

The total  $2^1S_0$  metastable beam entering the magnet spacer aperture,  $J(2^1S_0)$ , was approximately  $5 \times 10^8$  atoms/sec, as determined from the electron signal in combination with the electron-ejection efficiencies for gold and an assumed singlet-to-triplet metastable ratio in the beam of 1:3. With this atomic beam of metastable atoms (but no radiation incident), a steady background counting rate of 30-40 counts/sec was observed at the channeltron. If the helium gas was allowed to flow into the source chamber but the electron-bombarder collector current reduced to zero, the counting rate was decreased to dark current. The interruption of the metastable beam with a beam flag also reduced the counting rate to dark current; as the beam was well collimated, this implied the background resulted from interactions within the magnet spacer.

As the  $2^1P_1$  state (and other higher states) could

be excited by electron impact both directly and by cascades, this background was expected to at least partially arise from the scattering of 584-Å photons from the electron bombarder by residual helium gas in the magnet region. In the first of two experiments on the photon background, the magnitude was indirectly studied through the ratio of signal to background, where signal here refers to the 584-Å photons resulting from the ( $2^1S_0-2^1P_1$ ) excitation with a helium lamp. This ratio, typically 10:1 (Sec. VA), would be expected to increase if the photon contribution to the background was removed but the radiation incident on the beam unchanged. Any photon background was eliminated by alternately pulsing the electron bombarder on and off and observing the channeltron signal during the off period. A 2-kHz repetition rate was used, based on the 0.3-msec transit time of an atom over the 40-cm distance between the electron bombarder and the magnet. A pulse width of 0.2 msec gave optimal signal to background, although the duty cycle was not critical. A dual pulse counter was synchronously triggered so that the channeltron signal could be observed during both the on and the off period of the bombarder.

The ratio of signal to background was increased by a factor of 2 for the bombarder off period and decreased by a factor of 2 for the on period. The fractional contribution to the background from scattered photons may be estimated to be about 20 counts/sec from this result, assuming that the change in signal to background arose only from the elimination of photons. Also, from this result one can estimate a 584-Å photon current into the magnet spacer of  $6 \times 10^7$  photons/sec.

In the second background experiment, the magnetic field dependence of the background was determined; the results of about 6 h of signal averaging are shown in Fig. 7. The shape of the curve was that for a level-crossing signal when 584-Å photons were incident along the direction of the beam ( $\theta = \pi/2, \phi = -\pi/2$ ) and the scattered photons were observed by the channeltron ( $\theta' = \pi/2, \phi' = \pi$ ). A least-squares value obtained for the half half-width  $H_{1/2}(2^1P_1)$  was  $(94 \pm 36)$  G in good agreement with the theoretical value expected for 584-Å radiation (102.35 G).

The magnetic-field-dependent signal, however, was quite small compared to the total constant signal, i. e., about 3%, whereas in the first background experiment, the channeltron signal was estimated to be half due to photons. Since the magnetic-field-dependent ( $\alpha = \pi/2$ )-to-independent ( $\alpha = 0$ ) contributions to the photon scattering were in the ratio of 1:2 for this geometry (Sec. IIIA), one infers that the 584-Å radiation from the electron bombarder was polarized, with an intensity ratio of the components parallel ( $\alpha = 0$ ) and per-

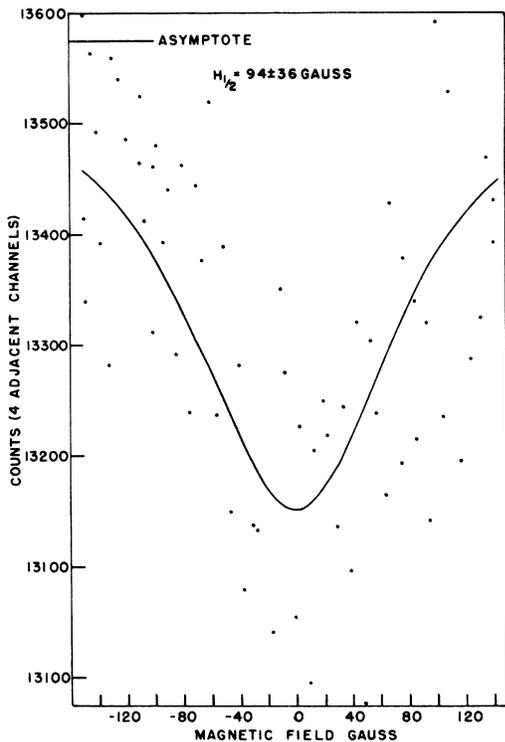


FIG. 7. Level-crossing signal from electron-bombarder photons. Solid curve and half half-width were calculated from least squares.

pendicular ( $\alpha = \pi/2$ ) to the magnetic field equal to about 8. Such strongly polarized radiation could be produced in the electron bombarder because the electron current and constraining magnetic field were parallel; when the direction of momentum transfer is along the quantization axis (magnetic field), only the ( $2^1P_1, m_J = 0$ ) state is excited.<sup>22</sup> As the electron bombarder and level-crossing magnetic fields were parallel, the principal ( $2^1P_1, m_J = 0$ )  $\rightarrow$   $1^1S_0$  decay radiation had the  $\alpha = 0$  polarization.

The possibility that the remaining nonphoton background originated from the  $2^1S_0$  metastable current of the beam was investigated by observing the channeltron signal when the beam was irradiated with a He<sup>4</sup> Osram lamp at the window between the electron bombarder and the magnet (Fig. 2). Although about one-third of the  $2^1S_0$  component of the beam was quenched in this manner, no statistically significant change in the background counting rate could be observed. This result implies that the remaining background did not primarily originate from two-photon free decay of the  $2^1S_0$  metastables in the beam although a crude estimate based on the  $2^1S_0$  radiative lifetime<sup>23</sup> (19.5 msec<sup>3</sup>) would indicate some effect might be observed. The source of this remaining background, whether  $2^1S_0$  free decay or

possible scattering processes involving both singlet and triplet metastables, is at present not clear.

## V. MEASUREMENTS

### A. $2^1P_1$ Lifetime

In the first measurements of the  $2^1P_1$  lifetime, He<sup>4</sup> resonance lamps were used. For the optical arrangement employed but with no infrared polarizer (see below), the fractional deexcitation of the  $2^1S_0$  beam with 20582-Å resonance radiation was about 0.09, as observed with the electrometer. Since the metastable atoms excited to the  $2^1P_1$  state decayed almost entirely to the  $1^1S_0$  ground state, the rates for the production of  $2^1P_1$  atoms and 584-Å photons were equal. For the total  $2^1S_0$  metastable beam  $J(2^1S_0)$  of approximately  $5 \times 10^8$  atoms/sec (Sec. IV), this production rate of 584-Å photons was about  $5 \times 10^7$  photons/sec. For the channeltron fractional solid angle and efficiency, the expected detector signal is hence about 1600 counts/sec. In practice, a linear infrared polarizer (type HR, Polaroid Corp., Cambridge, Mass.) was also used with its pass direction perpendicular to the magnetic field ( $\alpha = \pi/2$ ), since the incident radiation with the orthogonal polarization ( $\alpha = 0$ ) essentially contributed a constant 584-Å decay-photon signal. This polarizer, opaque in the visible, had a transmission of 75% at  $2 \mu$  in the pass direction or a total transmission of about one-third. With this polarizer, typical operating signals of 300–500 counts/sec were obtained, in accord with the above discussion. The signal (0 G) to background was hence about 10:1. Parenthetically, a total (into  $4\pi$  steradian) resonance-lamp output at 20582 Å of 20 mW is implied by the observed fractional deexcitation for an estimated  $2^1P_1$  excitation cross section [Eq. (7)] of  $5.4 \times 10^{-12}$  cm<sup>2</sup> (see also Sec. III D).

By observing the level-crossing signal for

$$|H| \leq 1.5H_{1/2}(2^1P_1) = 154 \text{ G},$$

the Breit-Franken formula (for a white incident profile) could be approximately used, with a possible systematic error of the order of several percent (Sec. III D). No effects from self-reversal were expected in light of the observed profiles of the resonance lamps.<sup>4</sup> Although of no real importance for these measurements, the effect of the 584-Å photons originating in the electron bombarder can be seen to be included in the assumed functional form for the level-crossing signal.

To take this data with the CAT, 200 sweeps were typically made in a run for a total signal-accumulation time of a little over an hour; a typical curve is shown in Fig. 8. This result and similar others were consistent with the theoretical value, for a purely radiative lifetime, of  $H_{1/2}(2^1P_1)$ . A similar result was recently obtained<sup>3</sup> with a He<sup>4</sup> resonance

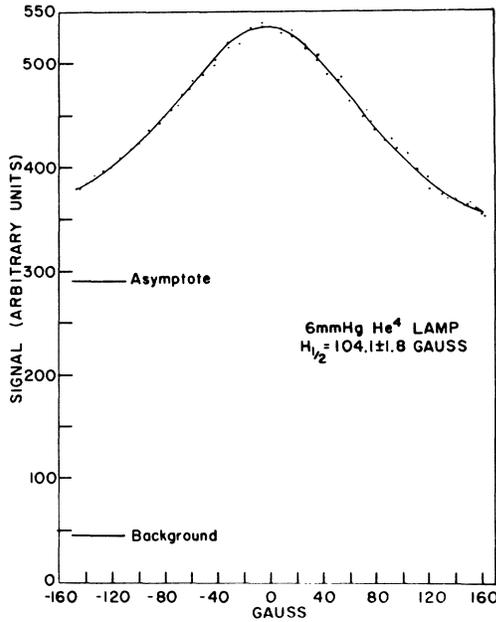


FIG. 8.  $2^1P_1$  lifetime level-crossing signal with helium resonance lamp. Solid curve and half half-width were calculated from least squares.

lamp, where, in addition,  $H_{1/2}(2^1P_1)$  was analyzed over different ranges of magnetic field; a significant line-profile influence on  $H_{1/2}(2^1P_1)$  was found when fields greater than 200 G were included. In the present study, a pronounced lamp-profile effect has been observed when comparatively large fields, e.g., 500 or 1000 G, are applied. Over the range of He<sup>4</sup>-lamp-filling pressures investigated, 1- to 6-mm Hg, no variation in the measured lifetime was observed.

Whereas the parameters  $G_2$ ,  $G_3$ , and  $G_4$  also obtained from the least-squares fit were consistent with expectations, the behavior of  $G_3 = \tan 2\phi$  was confirmed by investigating the effect of noncollinearity of incident and exiting radiation. In measurements similar to the above, two successive runs were made under conditions otherwise identical except for different angles of incidence ( $\phi$ ) obtained by tilting the optics. For approximate tilt angles of  $-4^\circ$  and  $+4^\circ$ ,  $G_3$  implied angles  $\phi$  of  $(-3.0 \pm 0.2)$  and  $(4.4 \pm 0.2)$  degrees, respectively. The other parameters, including the lifetimes, were consistent with expectations.

The difficulty arising from the resonance-lamp profile was completely circumvented by the use of a blackbody source of 20 582-Å radiation, although this was done at the expense of about an order of magnitude in signal. This observed signal is consistent with the expected intensity of the blackbody source. Using straightforward techniques,<sup>20</sup> an effective excitation cross section  $\sigma_{bb}$  for a black-

body source can be derived in a manner similar to that used for Eq. (7):

$$\sigma_{bb} = \pi r_0 c f [I(\nu_0)/\sigma T^4], \quad (9)$$

where the quantities  $r_0$ ,  $c$ , and  $f$  have the same meaning, and where  $[I(\nu_0)/\sigma T^4]$  is the fractional power per unit frequency radiated by the blackbody at  $\nu_0$  ( $\lambda_0 = 20\,582\text{ \AA}$ ). From the blackbody curves, we get

$$\sigma_{bb}(2^1S_0 - 2^1P_1) = 3 \times 10^{-17} \text{ cm}^2$$

for an assumed lamp temperature of 3000 °C (850-W input). As the experimental geometry was the same for the sun-gun and the He<sup>4</sup> resonance lamps, except for the germanium filter, the channeltron signal for the two sources was directly proportional only to lamp power and effective excitation cross section. Hence, the expected ratio of 584-Å-photon signals with blackbody and resonance-lamp excitation is

$$\frac{S_{bb}}{S} = (0.85) \frac{850}{(20 \times 10^{-3})} \frac{3 \times 10^{-17}}{(5.4 \times 10^{-12})} = 0.2,$$

where (0.85) accounts for the germanium filter transmission. With the same infrared linear polarizer, the observed 584-Å-photon signal (at 0 G) was of the order of 40 counts/sec with a corresponding signal-to-background ratio of about 1:1, in reasonable agreement with the above estimate in view of its approximate nature.

In a run, about 900 sweeps were made with the CAT for a data accumulation time of 5 h. The sweeping was concentrated on the region where the strongest magnetic field dependence of the signal occurred; typically, the signal was measured out to fields slightly greater than  $2H_{1/2}(2^1P_1)$ . However, observations of the 584-Å-photon signal with the pulse counter at high fields ( $\sim 1000$  G) agreed with the prediction of the Breit-Franken formula. A typical result is shown in Fig. 9. In all cases, the parameters  $G_2$  and  $G_4$  yielded the 1:1 signal-to-background ratio and  $G_3$  implied small angles ( $\phi$ ) of 1–2 degrees (as expected from the optics). In the data analysis, the signals in adjacent CAT channels were added (i.e., channel 0 to channel 1, channel 2 to channel 3, etc.), and the magnetic fields corresponding to each of the two adjacent channels averaged. Errors resulting from sweeping over a small range of magnetic field in each CAT channel were completely negligible; this was confirmed by comparing the average value of a Lorentzian about a field point with the value of the Lorentzian at that point, where the half half-width  $H_{1/2}(2^1P_1)$  and the range of field in two adjacent channels ( $\sim 6$  G) were used. Moreover, as previously mentioned, the level-crossing signal from the electron-bombarder photons was automatically included in the functional form analyzed. The error

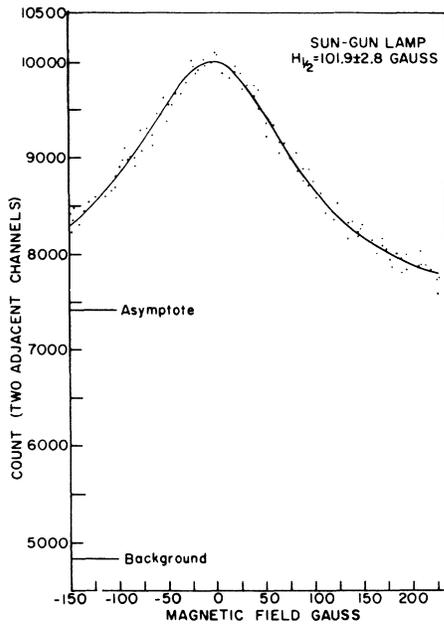


FIG. 9.  $2^1P_1$  lifetime level-crossing data with black-body excitation. Solid curve and half half-width were calculated from least squares.

in magnetic field measurement was estimated as 1%.

The half half-width obtained from these measurements was  $101.7 \pm 1.6$  G, where the uncertainty represents one standard deviation.<sup>24</sup> To this value the small correction of 0.4% for coherence narrowing must be made; a conservative error of 50% in the estimate of this effect has been allowed. With the inclusion of the 1% magnetic field uncertainty, the final experimental radiative half half-width is

$$H_{1/2}(2^1P_1) = 102.1 \pm 2.8 \text{ G.}$$

#### B. $3^1P_1$ Lifetime

The procedure for the measurement of the  $3^1P_1$  lifetime was, in principle, the same as for the  $2^1P_1$  state, although a number of modifications of technique were necessary. In order to selectively excite the beam with 5016-Å radiation ( $2^1S_0-3^1P_1$ ), an interference filter could have been used but at a sacrifice of intensity. However, as water is completely transparent at 5016 Å and completely opaque at  $2 \mu$ , a 2.5-cm-thick water filter was instead used to block the much more intense 20582-Å radiation. A Corning 3-73 glass filter, opaque below  $0.41 \mu$  and with a transmission of 86% at 5016 Å, was also used, both to prevent any ( $2^1S_0-4^1P_1$ ) excitation at 3964 Å and to eliminate the small channeltron signal due to its slight sensitivity to the shorter-wavelength radiation from the helium resonance lamps.

For the operating conditions in the  $2^1P_1$  lifetime work and with the same optical system (except no polarizer), the observed 537-Å-photon signal resulting from the 5016-Å excitation with a He<sup>4</sup> resonance lamp was about 10–15 counts/sec. Since the cross section [Eq. (7)] for ( $2^1S_0-3^1P_1$ ) excitation is one-tenth that for the ( $2^1S_0-2^1P_1$ ) transition, the observed 537-Å-photon signal is consistent with the 584-Å-photon signal, if the 5016-Å intensity is assumed to be one-tenth that of the 20582-Å radiation.

With a background of 30–40 counts/sec, the 537-Å-photon signal (0 G)-to-background ratio was roughly one-third. As only one-quarter of this signal was magnetic field dependent (no polarizer, Sec. III A), the useful level-crossing signal equalled 0.08 times the background. However, the 584-Å photons originating in the electron bombarder gave rise to a magnetic-field-dependent signal equal to 0.03 times the background. In light of these magnitudes, considerable error in the  $3^1P_1$  lifetime would be expected in an analysis of the combined signal based on a single half-width parameter ( $G_1$ ). Since the 584-Å-photon signal increased toward fields away from 0 G, it had the effect of decreasing the observed half half-width  $H_{1/2}(3^1P_1)$ . This was indeed observed in some preliminary data, where half half-widths obtained were too narrow by 10–20%. A least-squares fit with a single half-width parameter to a hypothetical sum of 537- and 584-Å signals of the above estimated relative magnitude implied this observed narrowing.

The signal from the 584-Å photons was eliminated by pulsing the electron bombarder on and off and observing while it was off [an inhibit pulse to the single-channel analyzer (Fig. 5) during the on period]. At the rate of 2 kHz, the electron bombarder was pulsed many times during the dwell time at a CAT address. Although this gating procedure doubled the ratio of signal to background, the signal amplitude was decreased by about a factor of 3, as roughly expected from the electron-bombardment duty cycle. To increase the signal, the helium resonance lamps were operated at about three times the power used in the  $2^1P_1$  lifetime work, and a signal to background of about 1:1 obtained. Since coherence narrowing effects were quite small, the gas flow into the source chamber was doubled with a corresponding increase in the interaction chamber pressure by a factor of 2. For these operating conditions a 537-Å-photon signal of 10–15 counts/sec was observed.

In these measurements, the data-accumulation time (5 h) and sweep rate were identical to those in the sun-gun work, and the signal was again measured in the region of greatest field dependence, i. e., a little further out than  $2H_{1/2}(3^1P_1)$ . A typical result is shown in Fig. 10. In the data analy-

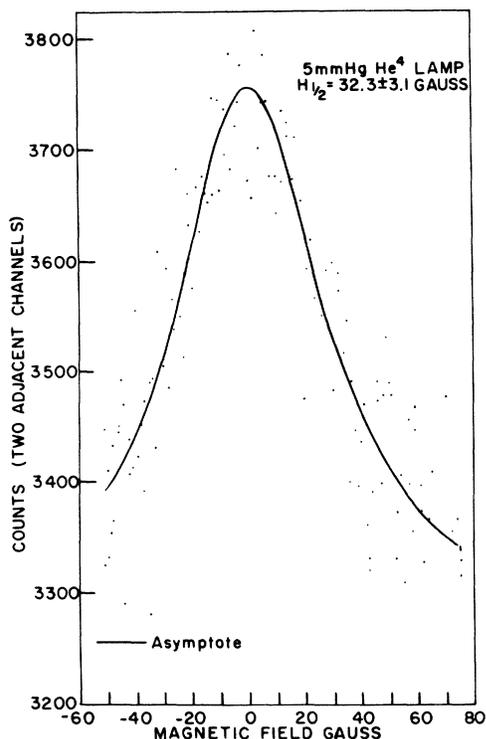


FIG. 10.  $3^1P_1$  lifetime level-crossing data. The solid curve and half half-width were calculated from least squares.

sis, counts in adjacent channels were again added. The parameters  $G_2$  and  $G_4$  implied the correct signal-to-background ratio (1:1), where  $G_4$  now included the signal from the  $\alpha=0$  polarization.  $G_3$  again implied angles of noncolinearity ( $\phi$ ) of 1 or 2 degrees. No magnetic field dependence was observed in the background during a test run of 3 h. The half half-width obtained from these measurements was  $32.8 \pm 1.8$  G, where the error is one standard deviation. With the allowance again of 1% uncertainty in magnetic field, the final experimental radiative half half-width is

$$H_{1/2}(3^1P_1) = 32.8 \pm 2.1 \text{ G.}$$

## VI. SUMMARY AND DISCUSSION

The present experimental values for the lifetime of the  $2^1P_1$  and  $3^1P_1$  states are in excellent agreement with theory.<sup>2</sup> The experimental together with the theoretical lifetimes are

$$\begin{aligned} \text{expt: } \tau(2^1P_1) &= (5.57 \pm 0.15) \times 10^{-10} \text{ sec,} \\ \tau(3^1P_1) &= (1.73 \pm 0.11) \times 10^{-9} \text{ sec;} \\ \text{theor: } \tau(2^1P_1) &= 5.555 \times 10^{-10} \text{ sec,} \\ \tau(3^1P_1) &= 1.726 \times 10^{-9} \text{ sec.} \end{aligned}$$

From these experimental lifetimes, measured

values for the absorption oscillator strengths,  $f(2^1P_1-1^1S_0)$  and  $f(3^1P_1-1^1S_0)$ , may be deduced. In the case of the  $2^1P_1$  state, a measured value for the oscillator strength can be obtained directly from the observed lifetime as the calculated ( $2^1P_1-1^1S_0$ ) to ( $2^1P_1-2^1S_0$ ) branching ratio is  $\sim 10^3$ . In the case of the  $3^1P_1$  state, the small correction for the ( $3^1P_1-2^1S_0$ ) decay can be made using the theoretical oscillator strength  $f(3^1P_1-2^1S_0) = 0.1514 \pm 0.0002$  which has been experimentally verified to several percent.<sup>25</sup> [The calculated ( $3^1P_1-1^1S_0$ ) to ( $3^1P_1-2^1S_0$ ) branching ratio is 42.] Hence, one has the theoretical and experimental oscillator strengths

$$\begin{aligned} \text{expt: } f(2^1P_1-1^1S_0) &= 0.275 \pm 0.007, \\ f(3^1P_1-1^1S_0) &= 0.073 \pm 0.005; \\ \text{theor: } f(2^1P_1-1^1S_0) &= 0.27616 \pm 0.00001, \\ f(3^1P_1-1^1S_0) &= 0.0734 \pm 0.0001. \end{aligned}$$

Despite the quoted accuracy of the theoretical oscillator strengths, the present level-crossing study (and a concurrent one<sup>3</sup>) were motivated by a lack of convincing experimental confirmation, especially in light of one discrepancy of 30%.<sup>18</sup> In the case of this discrepancy, the oscillator strength  $f(2^1P_1-1^1S_0)$  was determined from a linear extrapolation to zero pressure of the Lorentzian component of the width of the 7281-Å ( $3^1S_0-2^1P_1$ ) line from a resonance lamp. The slope of this width however was in good agreement (6%) with existing pressure-broadening theories when the theoretical oscillator strength was used.<sup>26</sup> In a study of emission profiles from an atomic beam,<sup>27</sup> where large calculated Doppler corrections were necessary, a value based on the most reliable line measured disagreed slightly with theory, whereas an average of four lines did agree. A number of recent experimental values for  $f(2^1P_1-1^1S_0)$  are summarized in Table I. The most recent measurements<sup>3, 30</sup> are all in agreement with theory. The present value is the most precise to date, although the estimated theoretical accuracy is still 700 times greater.

With regard to the  $3^1P_1$  lifetime, past experimental values have shown considerable scatter and frequently poor agreement with theory. In the emission profile study discussed above,<sup>27</sup> the theoretical value lay just outside the quoted error (less than in the present work). A recent value from beam-foil work is of comparable precision to the present experiment and both are in excellent agreement. A number of recent experimental values for the  $3^1P_1$  lifetime are summarized in Table II. With reference to the oscillator strength  $f(3^1P_1-1^1S_0)$ , the estimated precision of the theoretical value is a factor of 50 times greater than that of the present experiment.

In conclusion, a new level-crossing technique

