Lifetime of the First ¹P₁ State of Zinc, Calcium, and Strontium*

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The lifetime of the first excited ${}^{1}P_{1}$ state of zinc, calcium, and strontium has been measured by the zero-field level-crossing technique. The results are: zinc $\tau({}^{1}P_{1})=1.41\pm0.04\times10^{-9}$ sec, calcium $\tau({}^{1}P_{1})=4.48\pm0.15\times10^{-9}$ sec, and strontium $\tau({}^{1}P_{1})=4.97\pm0.15\times10^{-9}$ sec. The results are compared to earlier experimental results and also to theoretical predictions. It is found that our results are in excellent agreement with the Bates and Damgaard Coulomb approximation method for estimating the oscillator strengths.

I. INTRODUCTION

HE problem of determining atomic transition probabilities and excited-state lifetimes is an old one in physics, although it has received relatively little experimental attention in recent years. At the same time, advances in photoelectric recording and optical techniques make many early methods well worth reviewing and revising. Surveys of existing literature show a few advances in techniques since the early 1930's, while revealing upon occasion a rather considerable state of disagreement-not uncommonly by an order of magnitude or more—in many "absolute" measurements. Several excellent compilations of previous literature, tables of numerical values, and review articles exist. The best reference for all of these as well as individual articles, up to 1962, is the National Bureau of Standards monograph by Glennon and Wiese.1

Most absolute experimental determinations of oscillator strengths or transition probabilities of spectral lines have used the "hook" method developed by Rozhdestvenski and co-workers at Leningrad over the years from 1912 to the present.² This method and the various line absorption methods of measuring absolute oscillator strengths all require a knowledge of the density of the scattering atoms.³ It is this requirement that presents a limitation to the precision of these methods. (For the measurement of *relative* oscillator strengths, however, the "hook" method seems to be excellent.) The zero-field level crossing or Hanle effect,⁴

on the other hand, does not suffer from this requirement in the limit of low vapor densities, and should therefore be much more reliable. The level-crossing technique does require the production of strong unself-reversed resonance lines and consequently is limited to measurements of the oscillator strengths of the first few resonance lines of an element. In this respect the "hook" method would seem superior since it does not require resonance lines.

In a previous paper⁵ the measurement of the lifetime of the first excited 1P_1 state of Cd by the zero-field level-crossing technique is reported. In the present paper we report on the lifetime of the first excited 1P_1 state of Ca, Sr, and Zn. In these experiments the resonance radiation is scattered from an atomic beam so that the difficulty of finding a transparent cell which is not attacked by Ca or Sr vapor is avoided. Our results are compared to previous measurements and to theoretical calculations for the oscillator strengths and are found to agree surprisingly well with the Bates and Damgaard approximation.⁶

II. THEORY

We first present a simple and somewhat restricted theory derived on strictly classical grounds, to give as directly as possible a physical picture of the Hanle or zero-field level-crossing effect. Later, a more generalized quantum-mechanical treatment is given leading to the same conclusions.

More than one geometrical arrangement is possible, and polarized light need not be used, but the simplest arrangement conceptually is probably the following. Consider a vapor of the atoms to be investigated—either in the form of a free atomic beam, or in a closed evacuated container—with the scattering region at the coordinate origin (see Fig. 1). A beam of resonance radiation with the E field polarized in the Y direction is sent in along the X axis. Absorption of the resonant light by the vapor raises the atoms to the excited state whose lifetime is to be measured. Classically one may consider the resonance light to excite electric dipoles

214 (1925).

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¹B. M. Glennon and W. L. Wiese, Natl. Bur. Std. (U. S.), Monograph No. 50 (1962).

² See various articles in *Optical Transition Probabilities*, a collection of translated Russian articles published by the National Science Foundation, Washington, D. C., 1962.

⁸ A. C. G. Mitchell and M. W. Zemansky, Resonance Radiation and Excited Atoms (Cambridge University Press, New York, 1961). ⁴ W. Hanle, Z. Physik 30, 93 (1924); Ergeb. Exakt. Naturw. 4,

A. Lurio and R. Novick, Phys. Rev. 134, A608 (1964).
 D. R. Bates and A. Damgaard, Phil. Trans. 242, 101 (1949).

oscillating along the direction of the Y axis. These dipoles then re-radiate energy with the usual dipole radiation pattern, producing maximum intensity perpendicular to, and zero intensity along, the Y axis. However, if a magnetic field is present, aligned with the Z axis, the dipoles will precess about this field at their Larmor frequency ω until they re-radiate. ω depends on the field strength H via the Larmor relation $\omega = \gamma H$ where γ is the gyromagnetic ratio: in this case

$$\gamma = \omega/H = g_J \mu_0/\hbar \,, \tag{1}$$

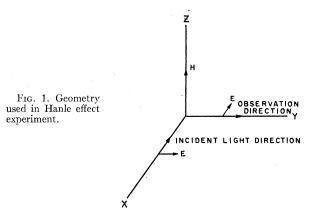
with g_J = the electronic g factor and μ_0 = the Bohr magneton. The angle of rotation through which a given dipole precesses before re-radiating is thus dependent on H. Multiplying the \sin^2 dipole-radiation pattern by an exponential damping factor to account for the random finite excited-state lifetimes over a large continuously excited collection of atoms, and integrating over all times, yields the following results for the observed intensity of scattered resonant radiation along the Y axis (at 90° to the incident beam) as a function of the magnetic field strength H:

$$I = C \int_0^\infty e^{-\Gamma t} \sin^2 \omega t dt = \frac{C}{2} \left[1 - \frac{\Gamma^2}{\Gamma^2 + 4\gamma^2 H^2} \right], \quad (2)$$

where Γ is the reciprocal of the mean life of the excited state and C is an arbitrary constant relating to the incident light intensity, the experimental geometry, and the number of scatterers. The result is just an inverted Lorentzian line shape as a function of H, with a full width at half-maximum equal to Γ . The half-maximum is seen to occur at a field value $H_{1/2}$ for which $4\gamma^2H_{1/2}^2=\Gamma^2$, or using Eq. 1, when

$$\Gamma = (2g_J \mu_0 H_{1/2}/\hbar) \text{ sec}^{-1}.$$
 (3)

Thus, a simple determination of the value of the magnetic field required to produce the half-maximum level in the Lorentzian line shape serves in principle to determine the mean life $\tau=1/\Gamma$, since g_J is generally well known. In practice fairly detailed profiles of the scattered intensity were taken as a function of H, as described in the next section, in order to carefully analyze and verify the Lorentzian shape. If the incident light along the X axis is unpolarized, one may carry out the theoretical analysis by resolving this light into two orthogonal polarizations. For the component polarized parallel to the Y axis the treatment is the same as given above. For the component polarized parallel to the Z axis, the scattered light is independent of the applied magnetic field since a dipole oscillating parallel to H does not precess. In practice one polarizes the incident light parallel to the Y axis in order to eliminate this constant background.



In odd A isotopes the presence of hyperfine structure may alter the Lorentzian profile in particular cases. This occurs since the g_F value of the odd isotopes will differ from the g_J value for the even isotopes. Moreover, the Hanle effect is only a special case at zero field of the more general "level-crossing" phenomenon discovered by Franken and co-workers⁸ and the observational technique used here will also detect these nonzero-field crossings. This may then introduce distortions in the wings of the Lorentzian profile if fields large enough to reach nonzero-field level crossings are used, which may be the case with comparatively small hyperfine intervals and short lifetimes (e.g., $\tau \le 10^{-9}$ sec). This was not of importance for the elements studied here since the naturally occuring group II elements contain at most small amounts of odd A isotopes and those odd A isotopes which do occur all have $I \ge \frac{5}{2}$. Thus for these isotopes the level-crossing signals are a very small fraction of the scattered resonance radiation because of the large number of Zeeman levels.

We shall now give a quantum-mechanical derivation for the intensity of the scattered resonance radiation from a collection of identical atoms with I=0, excited from the ${}^{1}S_{0}$ ground state to the ${}^{1}P_{1}$ state and decaying back to the ${}^{1}S_{0}$ state. We will take the externally applied static magnetic field to be along the Z axis but will allow for arbitrary incident light and observing directions (see Fig. 2). The general expression for the rate of scattering of resonance radiation has been given by Breit⁹ and Franken⁸ and is

$$R(f,g) \propto N \sum_{\substack{\mu\mu' \\ mm'}} \frac{f_{\mu m} f_{m\mu'} g_{\mu'm'} g_{m'\mu}}{\Gamma - i \left[\left(E_{\mu} - E_{\mu'} \right) / \hbar \right]}, \tag{4}$$

where $f_{\mu m} = (\mu | \hat{e} \cdot \mathbf{r} | m)$, $g_{\mu m} = (\mu | \hat{e}' \cdot \mathbf{r}' | m)$, μ and μ' refer to the excited state magnetic sublevels, m and m' refer to the ground-state sublevels, and E_{μ} is the energy of the μ th excited state. Figure 2 shows the spherical

⁷ The classical treatment gives the correct results for the simple case of the excitation from a J=0 ground state to a J=1 excited state. We have not examined the classical treatment in more complicated cases.

⁸ P. Franken, Phys. Rev. 121, 508 (1961).
⁹ G. Breit, Rev. Mod. Phys. 5, 91 (1933).

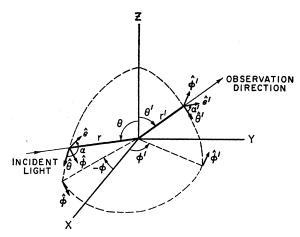


Fig. 2. Coordinate and polarization system used in quantum mechanical calculation of zero-field level-crossing signal.

coordinate system used to evaluate R. The incident light is directed toward the origin at coordinate angles θ and φ and the light polarization vector \hat{e} makes an angle α with respect to θ and lies in a plane normal to \mathbf{r} . \hat{e}' and \mathbf{r}' refer to the scattered light. With this coordinate system we have

 $\hat{e} = \hat{\imath}(\cos\theta\cos\varphi\cos\alpha - \sin\varphi\sin\alpha)$

$$+\hat{\jmath}(\cos\theta\sin\varphi\cos\alpha+\cos\varphi\sin\alpha)-\hat{k}(\sin\theta\cos\alpha).$$
 (5)

We may define a new set of complex vectors which will

prove useful by the relations

$$e_{\pm} = e_x \pm i e_y = (\cos\theta \cos\alpha \pm i \sin\alpha) e^{\pm i \varphi},$$

$$e_z = -\sin\theta \cos\alpha,$$

$$r_{+} = r_x \pm i r_y,$$
(6)

so that $\hat{e} \cdot \mathbf{r} = \frac{1}{2}(e_+r_- + e_-r_+) + e_zr_z$. In order to evaluate $f_{m\mu}$ and $g_{m\mu}$ we need the matrix elements¹⁰

$$\langle \gamma, 0, 0 | e_{\pm} r_{\mp} | \gamma', 1, \pm 1 \rangle = \mp \sqrt{2} e_{\pm} \langle r \rangle, \langle \gamma, 0, 0 | e_{z} r_{z} | \gamma', 1, 0 \rangle = e_{z} \langle r \rangle,$$
(7)

where $\langle r \rangle$ is the reduced matrix element of **r**. Let us now define the excitation matrix $F_{\mu\mu'} = \sum_m f_{\mu m} f_{m\mu'}$ or more explicitly

$$F_{\mu\mu'} = \begin{vmatrix} \frac{1}{2}(e_{+}e_{-}) & -e_{-}e_{z}/\sqrt{2} & -\frac{1}{2}e_{-}^{2} \\ -e_{+}e_{z}/\sqrt{2} & e_{z}^{2} & e_{z}e_{-}/\sqrt{2} \\ -\frac{1}{2}e_{+}^{2} & e_{z}e_{+}/\sqrt{2} & \frac{1}{2}e_{+}e_{-} \end{vmatrix} |\langle r \rangle|^{2}. \quad (8)$$

For our simple case the re-radiation matrix $G_{\mu'\mu} = \sum_{m'} g_{\mu'm'}g_{m'\mu}$ is the same matrix as $F_{\mu\mu'}$ except that all quantities are primed since they refer to the scattered light.

It is convenient for understanding the coherent scattering process to break R up into different $\Delta\mu$ components where $\Delta\mu = |\mu - \mu'|$. Then

$$R(f,g) = R(\Delta \mu = 0) + R(\Delta \mu = 1) + R(\Delta \mu = 2)$$
, (9)

and

$$R(\Delta\mu = 0) = \sum_{\substack{\mu, \mu' \\ \mu = \mu'}} \frac{F_{\mu\mu'}G_{\mu'\mu}}{\Gamma - (i/\hbar)(E_{\mu} - E_{\mu'})} = \frac{1}{2\Gamma} (1 - e_z^2 - e_z'^2 + 3e_z^2 e_z'^2), \qquad (10)$$

$$R(\Delta\mu=1) = \sum_{\substack{\mu,\mu' \\ |\mu-\mu'|=1}} \frac{F_{\mu\mu'}G_{\mu'\mu}}{\Gamma-(i/\hbar)(E_{\mu}-E_{\mu'})} = \frac{\frac{1}{2}e_ze_z'}{\Gamma^2+(E_{1,1}-E_{1,0})^2/\hbar} \left[\Gamma(e_-e_+'+e_+e_-') + \frac{i(E_{1,1}-E_{1,0})}{\hbar}(e_-e_+'-e_+e_-')\right]$$

$$+\frac{\frac{1}{2}e_{z}e_{z}'}{\Gamma^{2}+(E_{1,0}-E_{1,1})^{2}/\hbar}\left[\Gamma(e_{-}e_{+}'+e_{+}e_{-}')+\frac{i(E_{1,0}-E_{1,-1})}{\hbar}(e_{-}e_{+}'-e_{+}e_{-}')\right], \quad (11)$$

$$R(\Delta \mu = 2) = \sum_{\substack{\mu, \mu' \\ |\mu - \mu'| = 2}} \frac{F_{\mu \mu'} G_{\mu' \mu}}{\Gamma - (i/\hbar) (E_{\mu} - E_{\mu'})} = \frac{1}{4 [\Gamma^2 + (E_{1,1} - E_{1,-1})^2/\hbar]}$$

$$\times \left[\Gamma(e_{-}^{2}e_{+}^{\prime 2} + e_{+}^{2}e_{-}^{\prime 2}) + \frac{i(E_{1,1} - E_{1,-1})}{\hbar} (e_{+}^{2}e_{-}^{\prime 2} - e_{-}^{2}e_{+}^{\prime 2}) \right]. \quad (12)$$

For our experimental arrangement $\theta = \theta' = \frac{1}{2}\pi$, $\varphi = 0$, $\varphi' = \frac{1}{2}\pi$. Also $E_{1,1} = -E_{1,-1} = g_J \mu_0 H$ and $E_{1,0} = 0$ so that we may simplify the above equations to

$$R(\Delta\mu=0) = (1/\Gamma)(\frac{1}{2}\sin^2\alpha\sin^2\alpha' + \cos^2\alpha\cos^2\alpha'), \quad (13)$$

$$R(\Delta \mu = 1) = \frac{1}{2} \frac{g_{J}\mu_{0}H \sin 2\alpha \sin 2\alpha'}{\Gamma^{2} + (g_{J}\mu_{0}H/\hbar)^{2}},$$
 (14)

$$R(\Delta \mu = 2) = -\frac{\Gamma/2 \sin^2 \alpha \sin^2 \alpha'}{\Gamma^2 + (2g_J \mu_0 H/\hbar)^2}.$$
 (15)

If the incident light is polarized with $\alpha = \frac{1}{2}\pi$ (i.e., parallel to the Y axis) and the outgoing intensity is

¹⁰ E. U. Condon and G. H. Shortley, *The Theory of Atomic Spectra* (Cambridge University Press, Cambridge, 1935), Chap. III.

measured without polarizers then

$$R(f,g) = \frac{1}{2} \left[\frac{1}{\Gamma} - \frac{\Gamma}{\Gamma^2 + (2g_J \mu_0 H/\hbar)^2} \right].$$
 (16)

To obtain Eq. (16), the result for the intensity of the scattered light, we have summed Eq. (9) for α' and for $\alpha' + \pi/2$ since this takes account of both orthogonal components of the outgoing beam.

Experimental Details and Results

In Fig. 3 is shown a schematic diagram of the apparatus. The beam source is a stainless-steel oven positioned about 4 in. below the scattering region. The beam effuses through a \frac{1}{4}-in.-diam crinkle foil aperture at the top of the oven and is further collimated into a $\frac{1}{2}$ -in.-diam region as it passes vertically through the scattering chamber. The beam is then collected on a liquid N_2 trap. A beam flag is provided to determine the degree of instrumental scattering in the absence of the beam and to aid in aligning the optics. Under actual conditions of operation the instrumental scattering can be reduced to several times the dark current of the photomultiplier while the resonance radiation scattered from the beam can easily be made 30 times the dark current. In this arrangement the dc scattered resonance radiation signal is collected by an aluminized light pipe, passed through a narrow-band interference filter for the ${}^{1}S_{0}$ — ${}^{1}P_{1}$ resonance line in order to reduce the scatteredlight background, and detected by a Dumont 7664 photomultiplier. The photomultiplier output is further amplified by a Hewlett Packard 412A vacuum-tube voltmeter and displayed on a servo recorder.

The externally applied vertical magnetic field at the scattering region was calibrated by proton resonance and found to be 7.51±0.02 G/A. The principal component of the stray magnetic field at the scattering region was 0.35 G in the vertical direction. This field was not compensated but was taken into account in evaluating

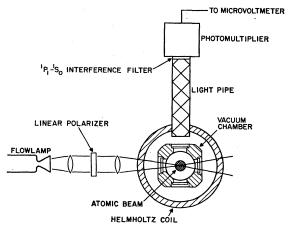


Fig. 3. Schematic diagram of apparatus.

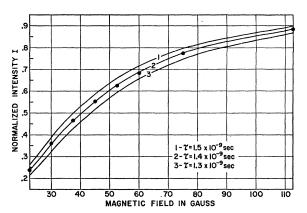


Fig. 4. Normalized plot of level-crossing signal versus magnetic field for zinc. 1, 2 and 3 are theoretical curves. Only the portion of the curve near the half-width is shown.

the data. Most of the measurements were made with the incident light polarized with the electric vector in the direction of observation. Measurements were also made with unpolarized incident light and gave the same results.

The data was taken in the following manner. By means of a reversing switch, the intensity I of the scattered light was recorded successively with the magnetic field in the "up" and then in the "down" direction. A series of about ten such determinations for values of H between 0 and 200 G comprised a run. A plot of Iversus H with the zero-field scattered-light intensity subtracted should be an inverted Lorentzian curve. From an approximate value of τ and the value of I at the larger magnetic-field values we may obtain the constant C in Eq. 2. For each run, after C has been determined, the experimental curve is normalized to unity amplitude and plotted together with a family of theoretical Lorentzian curves with different values of τ . In this way τ and an estimate of the uncertainty in τ can be obtained. Between 10 and 15 runs were taken for each element and the values for τ quoted below are the average values derived from these independent runs. Figures 4 and 5 show typical data for zinc and calcium. The slight asymmetry between the field up and the field down points for calcium is due to the incident and observing directions not being exactly perpendicular. Careful arrangement of the optics can eliminate this asymmetry. We find for the lifetime of the ¹P₁ states the following values: $\tau(\text{Zn}) = 1.41 \pm 0.04 \times 10^{-9}$ sec, $\tau(\text{Ca}) = 4.48 \pm 0.15 \times 10^{-9}$ sec, and $\tau(\text{Sr}) = 4.97$ $\pm 0.15 \times 10^{-9}$ sec. The quoted errors are twice the standard deviations for the different runs and represent a conservative estimate of the accumulated uncertainty arising from possible instrumental inaccuracies. We have taken $g_J = 1.00$ in obtaining τ from the measured linewidths.

The beam density was varied over a factor of about 20 during these measurements in order to detect and correct for coherence narrowing. At the highest beam

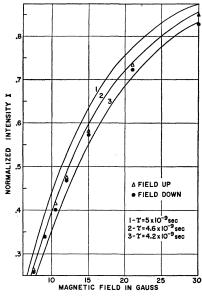


Fig. 5. Normalized plot of level-crossing signal versus magnetic field for calcium. 1, 2, and 3 are theoretical curves.

densities a narrowing of a few percent was obtained. For this reason the results quoted are from those runs taken at low-beam densities.

The possibility exists that self-reversal in the lamp might affect the results. One would expect this effect to distort the Lorentzian line shape. This would occur because as the magnetic field is increased at the scattering atoms, the $\Delta m = \pm 1$ Zeeman levels of the atom would scan a higher intensity part of the lamp profile, and consequently the scattered light would increase faster than it would for a flat profile. No such distortion of the measured Lorentzian shaped lines occurred to within the accuracy of the experiment.

Finally, we wish to mention that originally data for all

TABLE I. Zinc.

Author	Quantity measured	Result
Filippov ^a	$\tau(^{3}P_{*})/\tau(^{1}P_{*})$	6.76×10^{-5}
Auslander ^b	$\tau(^{1}P_{1})$	$2.3 \times 10^{-9} \text{ sec}$
Present work	$\tau(^1P_1)$	$1.41 \pm 0.04 \times 10^{-9} \text{ sec}$
Landmane	$\tau(^1P_1)$	$1.38 \pm 0.05 \times 10^{-9} \text{ sec}$
Billiter-Soleilletd	$\tau(^3P_1)$	$2.1 \times 10^{-5} \text{ sec}$
Byrone	$\tau(^3P_1)$	$2.0\pm0.2\times10^{-5}$ sec
Dumont ^f	$ au(^3P_1)$	$2.9-3.5\times10^{-5}$ sec

a A. N. Filippov, Physik Z. Sowjetunion 1, 289 (1932). See also Optical Transition Probabilities, a translation of selected Russian articles published for the National Science Foundation, Washington, D. C. by the Israel Program for Scientific Translations, Jerusalem, 1962. In this paper the ratio of the oscillator strengths is given and we have converted to the lifetime

ratio.

b J. Auslander, Helv. Phys. Acta 11, 562 (1938).

e A. Landman and R. Novick, Phys. Rev. 134, A56 (1964).

d W. Billeter, Helv. Phys. Acta 7, 841 (1934); P. Soleillet, Compt. Rend.

204, 253 (1937).

e F. W. Byron, M. N. McDermott, R. Novick, B. W. Perry, and E. Saloman, Phys. Rev. 134, A47 (1964).

f M. Dumont, Thèse de Troisième Cycle, Paris, 1962 (unpublished).

TABLE II. Calcium.

Author	Quantity measured	Result
Prokofev ^a	$\tau(^{3}P_{1})/\tau(^{1}P_{1})$	1.25×10 ⁻⁵
Steinhäuser ^b Ostrovskii <i>et al.</i> ^c	$ au^{(1}P_1) \ au^{(1}P_1)$	$3.5 \times 10^{-9} \text{ sec}$
Ostrovskii et at.	$\tau(^{1}P_{1})$ $\tau(^{1}P_{1})$	$6.2\pm1\times10^{-9}$ sec $5.40\pm0.14\times10^{-9}$ sec
Odintsove	$\tau^{(^1P_1)}$	$5.0\pm0.14\times10^{-9}$ sec
Present	$\tau^{(1}P_1)$	$4.48 \pm 0.15 \times 10^{-9} \text{ sec}$

three elements, with standard deviations of 3-6%, were taken with a somewhat cruder version of the present apparatus. The data presented here were obtained in a physically different location, with entirely different hardware, including different recording instruments and calibrating standards. The results presented here fall within the standard deviations of the preliminary results. This degree of care was taken in part because of disagreement between the preliminary data and previous values in the literature.

Discussion of Results

Tables I, II, and III list the present and previous results for the lifetime of the first excited ${}^{1}P_{1}$ and ${}^{3}P_{1}$ states of zinc, calcium, and strontium. These lifetimes have been calculated from the quoted f values by use of the relation³

$$f\tau = \frac{mc}{8\pi^2 e^2} \frac{g_2}{g_1} \lambda^2 = 1.499 \frac{g_2}{g_1} \lambda^2.$$

In the case of zinc, one has excellent agreement between the present results and those of Landman although it should be observed that both of these experiments employ the zero-field level-crossing technique. Moreover, the ratio of our $\tau({}^{1}P_{1})$ to the $\tau({}^{3}P_{1})$ of either Byron or Billiter-Soleillet is in excellent agreement with the ratio measured by Filipov. For the cases of calcium and strontium our $\tau({}^{1}P_{1})$ is lower than those of the

TABLE III. Strontium.

Author	Quantity	Result
Prokofev* Ostrovskii et al.b Ostrovskii and Penkino Present work	$ au^{(3P_1)/ au^{(1}P_1)} au^{(1P_1)} au^{(1P_1)} au^{(1P_1)} au^{(1P_1)} au^{(1P_1)}$	2.69×10^{-5} 6.38×10^{-9} sec 6.2×10^{-9} sec $4.97 \pm 0.15 \times 10^{-9}$ sec

<sup>V. K. Prokofev, Z. Physik 50, 701 (1928).
J. Ostrovskii, N. P. Penkin and L. N. Shabanova, Dokl. Akad. Nauk SSSR 120, 66 (1958) [English transl.: Soviet Physics—Doklady 3, 538 (1958)].
Yu. I. Ostrovskii and N. P. Penkin, Opt. i Spectroskopiya 11, 565 (1961) [English transl.: Opt. Spectry. (USSR) 11, 307 (1961)].</sup>

<sup>V. K. Prokofev, Z. Physik 50, 701 (1928).
A. Steinhäuser, Z. Physik 95, 669 (1935); 99, 300 (1936).
I. Ostrovskii, N. P. Penkin, L. N. Shabanova, Dokl. Akad. Nauk. SSSR 120, 66 (1958) [English transl.: Soviet Phys.—Doklady 3, 538 (1958)].
I. Ostrovskii, N. P. Penkin, Opt. i Spektroskopiya 11, 565 (1961) [English transl.: Opt. Spectry. (USSR) 11, 307 (1961)].
A. I. Odintsov, Opt. i. Spektroskopiya 14, 322 (1963) [English transl.: Opt. Spectry (USSR) 14, 172 (1963)].</sup>

TABLE IV. Intermediate coupling coefficients.

	Wolfe method	Lifetime ratio method
Zn	$c_1 = 0.5656$ $c_2 = 0.8247$	0.5657 0.8246
Srª	$c_1 = 0.5519$	0.5526
Caa	$ \begin{array}{cc} c_2 & 0.8339 \\ c_1 & 0.5709 \end{array} $	$0.8334 \\ 0.5717$
	$c_2 = 0.8210$	0.8204

 $[^]a$ We use the Prokofev lifetime ratio in the case of Ca and Sr since no direct measurement of $\tau^3P_1)$ is available for these elements.

Russian measurements, however, we note that their most recent results are in better agreement with ours. We do not know the source of these discrepancies but we should point out the following fact: The recent ingenious improvement in the hook method¹¹ utilizes total absorption to measure $Nf\Gamma$. The use of this method requires that the absorption follow a Lorentzian law at about three Doppler widths from the line center or typically about 200 times the natural linewidth from the line center. It is important to note that this has never been checked experimentally and any deviation from a Lorentzian line shape well out in the wings of the line could lead to erroneous results.

The excellent agreement in the case of Cd and Zn between the hook measurement of $\tau({}^{1}P_{1})/\tau({}^{3}P_{1})$ and the recent measurements of $\tau({}^{1}P_{1})$ and $\tau({}^{3}P_{1})$ for these states indicates that one may get good estimates of $\tau(^{3}P_{1})$ by using our $\tau(^{1}P_{1})$ and the ratio obtained from the hook measurements.

It is also of interest to compare the intermediate coupling coefficients¹² c_1 and c_2 for the ¹ P_1 and ³ P_1 states as derived from lifetime measurements with those derived from Wolfe's12,13 theory which makes use of the deviation of the fine structure from an interval rule. This theory takes account of spin-orbit and spin-otherorbit interactions but does not allow for spin-spin interactions or the fact that the ${}^{1}P_{1}$ and ${}^{3}P_{1}$ states can have slightly different radial wave functions. The values of c_1 and c_2 calculated by the two different methods are given in Table IV and are in very good agreement.

TABLE V. Comparison of theoretical and experimental values of the oscillator strengths.

	Ca	Sr	Zn
$\lambda({}^{1}S_{0}-{}^{1}P_{1})$	4227 Å	4608 Å	2139 Å
f (exp)a	1.80	1.92	1.46
$f \stackrel{.}{B} \stackrel{.}{\&} D)^{\mathrm{b}}$	1.83	1.90	1.49
f (Vainshtein)c	2.32		
f (Trefftz)d	1.737-1.223		

As pointed out in the introduction, the lifetime of the ${}^{1}P_{1}$ state provides a test of approximate methods used to calculate the oscillator strength of the ${}^{1}P_{1}$ to ${}^{1}S_{0}$ transition. The prediction of the Coulomb approximation method of Bates and Damgaard⁶ can be obtained by making use of the formula of Garstang.¹⁴

$$({}^{1}S_{0}||P||^{1}P_{1}) = -\sqrt{2}(c_{1}+\sqrt{2}c_{2})\sigma \cong -\sqrt{6}\sigma$$
,

and

$$S(J,J^1) = |-\sqrt{2}(c_1+\sqrt{2}c_2)\sigma|^2 \cong 6\sigma^2$$
,

where σ^2 is obtained for the p electron from the tables given in the Bates and Damgaard paper. The estimate of the oscillator strengths f from this calculation is shown in Table V. Also included in Table V are other estimates for f using explicit forms for the wave functions of the two states. The surprising conclusion to be drawn from Table V is that the Coulomb approximation is in very good agreement with the experimental results.

Note added in proof. After this article was submitted for publication a paper with the following reference appeared: E. Hulpke, E. Paul, and W. Paul, Z. Physik 177, 257 (1964). In this paper the following results are given: $\tau(Ca) = 4.67 \pm 0.11 \times 10^{-9}$ sec and $\tau(Sr) = 4.56$ $\pm 0.21 \times 10^{-9}$ sec, which are in excellent agreement with ours. As in their paper we have also considered the transitions from the ${}^{1}P_{1}$ to the ${}^{1}D_{2}$ state to be negligible compared to the decay to the ground state.

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