

Lifetime of the First Excited 1P_1 State of Mg and Ba; hfs of Ba^{137}

ALLEN LURIO

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

(Received 22 May 1964)

The lifetime of the first excited 1P_1 state of Mg and Ba has been obtained from the measured width of the zero-field level-crossing signal in the scattering of resonance radiation from an atomic beam of these elements. The results are $\tau(\text{Mg}) = (1.99 \pm 0.08) \times 10^{-9}$ sec and $\tau(\text{Ba}) = (8.2 \pm 0.2) \times 10^{-9}$ sec. These results are compared with previous measurements and in conjunction with other experimental results are used to obtain the intermediate coupling coefficients for the mixing of the 1P_1 and 3P_1 states. From a measurement of the one observable high-field level crossing in Ba^{137} , the dipole coupling constant for the $6s6p\ ^1P_1$ state is found to be -113.2 ± 1.0 Mc/sec in good agreement with the results of Jackson and Tuan. The quadrupole moment of the Ba^{137} is calculated to be 0.20×10^{-24} cm² from the data of Jackson and Tuan.

INTRODUCTION AND DESCRIPTION OF LIFETIME EXPERIMENTS

THE use of the level-crossing technique to measure the lifetime and hfs of the first excited 1P_1 state of the group II elements has been described previously.¹⁻³ We present here only a brief description of the lifetime measurement technique which is described more completely in Refs. 2 and 3. Barium and magnesium resonance radiation arising from transitions between the ground 1S_0 state and the first excited 1P_1 state (see Fig. 1) is incident on an atomic beam of the appropriate atoms (see Fig. 3 of Ref. 3). The radiation scattered at 90° to the incident light direction is detected by a photomultiplier whose dc output is recorded as a function of a static magnetic field applied to the scattering atoms in a direction perpendicular to the incident and observing directions. From the half-width of the Lorentzian-shaped zero-field level-crossing signal, the lifetime of the 1P_1 state is determined.

The resonance radiation was produced by either a flow lamp⁴ or a hollow cathode lamp. For Mg a flow lamp was used. For Ba it was found that the hollow cathode lamp produced much more stable though somewhat less intense resonance radiation and consequently the hollow cathode lamp was used in taking most of the data.

Metallic Ba and Mg were heated in stainless steel ovens to produce the beams of these elements. In some of the work, however, it was useful to have enriched isotopes of Ba in the beam. The enriched isotopes were supplied from Oak Ridge National Laboratory in the form of $BaCO_3$. In order to produce a Ba beam, finely powdered aluminum was mixed with the carbonate and heated in the stainless oven. It was found that a reaction, producing a Ba beam, started at about 700°C and that for steady operation a temperature in the region of 600°C was needed.

¹ A. Landman and R. Novick, Phys. Rev. **134**, A56 (1964).

² A. Lurio and R. Novick, Phys. Rev. **134**, A608 (1964).

³ A. Lurio, R. L. deZafra, and R. J. Goshen, Phys. Rev. **134**, A1198 (1964).

⁴ B. Budick, R. Novick, and A. Lurio, Appl. Optics (to be published).

LIFETIME RESULTS AND DISCUSSION OF RESULTS

A. Magnesium

Magnesium occurs naturally with 89.9% even isotopes ($I=0$) and 10.1% odd isotope. The odd isotope has a spin of $\frac{5}{2}$ so that it should have a negligible effect on the even isotope level-crossing signal (see Ref. 3). In order to obtain the lifetime, the experimental data were plotted against a family of theoretical level crossing curves. The result, the average of 12 runs, is $\tau = (1.99 \pm 0.08) \times 10^{-9}$ sec, where we have taken $g_J = 1.00$ for the 1P_1 state. These data were taken over a range of atomic beam densities and within the experimental uncertainty no multiple scattering narrowing of the level crossing width was observed.

Table I gives the present and previous values for the lifetime of the 1P_1 and the 3P_1 state. It is seen that our measured lifetime is shorter than those obtained previously. The disagreement with Ostrovskii *et al.* is not too surprising in view of the well-known difficulty of measuring absolute vapor densities which are necessary to obtain absolute oscillator strengths from anomalous dispersion experiments. Our disagreement with

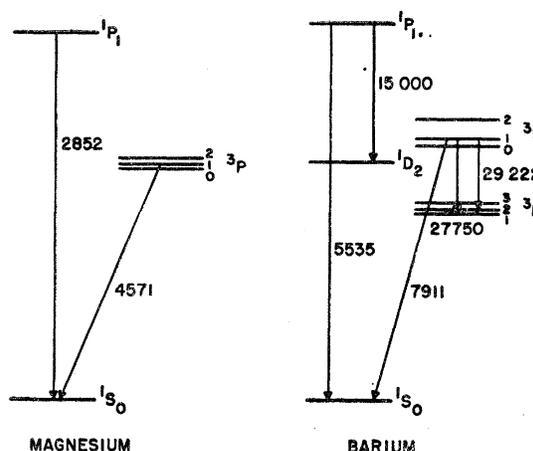


FIG. 1. Term level diagram for magnesium and barium. Wavelengths in Å.

TABLE I. Experimental and theoretical oscillator-strength values for $3s3p\ ^1P_1$ and 3P_1 states of magnesium.

Author	Lifetime (sec)	Oscillator strength
a. Experimental		
Ostrovskii <i>et al.</i> ^a		$f(^1P_1) = 1.2 \pm 0.3$
Demtroder ^b	$\tau(^1P_1) = (3.29 \pm 0.16) \times 10^{-9}$	$f(^1P_1) = 1.11 \pm 0.05$
Present work	$\tau(^1P_1) = (1.99 \pm 0.08) \times 10^{-9}$	$f(^1P_1) = 1.85 \pm 0.07$
Boldt ^c		$f(^3P_1) = 5.0 \times 10^{-6} \pm 20\%$
b. Theoretical		
Bates and Damgaard ^d		$f = 1.60$
Treffitz ^e		$f = 1.674$

^a Yu. I. Ostrovskii, N. P. Penkin, and L. N. Shabanova, Dokl. Akad. Nauk SSSR **120**, 66 (1958) [English transl.: Soviet Phys.—Doklady **3**, 538 (1958)].

^b W. Demtroder, Z. Physik **166**, 42 (1962).

^c G. Boldt, Z. Physik **150**, 205 (1958).

^d D. R. Bates and A. Damgaard, Phil. Trans. Roy. Soc. London **A242**, 101 (1950).

^e E. Treffitz, Z. Astrophys. **28**, 67 (1950).

Demtroder might be explained by radiation trapping in his experiment. It should be noted that he does not give his results as a function of vapor density as in the more recent work of Hulpke, Paul, and Paul, who used the same experimental method as Demtroder and who found a strong dependence of the lifetime on vapor density for elements with large oscillator strengths.

In Table III is shown the value of the intermediate coupling coefficients c_1 and c_2 for the mixing of the 3P_1 and 1P_1 states. We have computed them by the Wolfe method⁶ and by the lifetime ratio method⁵ where $\tau(^3P_1)$ is obtained from Boldt. While the values of c_1 and c_2 are both very close to the pure L - S coupling value and appear to agree rather well, the lifetime ratio $\tau(^3P_1)/\tau(^1P_1)$ would actually have to be reduced by about one-half in order to agree with the coupling coefficients obtained by the Wolfe method. However, it is likely that the small deviations from an interval rule for light elements such as Be and Mg cannot be accounted for by the simple theory of Wolfe and the more rigorous theory of Araki⁷ should be used.

B. Barium

Barium occurs naturally with 82.1% even isotopes and 17.9% odd isotopes both of which have spin $\frac{3}{2}$ (Ba^{135} , Ba^{137}). It was thought that the presence of 17.9% of odd isotopes might noticeably effect the shape of the even isotope level-crossing signal. For this reason a sample containing 98.0% of Ba^{138} and 1.6% of odd isotopes was obtained from Oak Ridge. It was found that to within the precision of the present experiment, the lifetime results of the natural and the enriched samples were identical. The Ba data were analyzed in the same manner as the magnesium with the result $\tau(^1P_1) = (8.20 \pm 0.25) \times 10^{-9}$ sec, where we have taken $g(^1P_1) = 1.025$.⁸

⁸ A. Lurio, M. Mandel, and R. Novick, Phys. Rev. **126**, 1758 (1962).

⁶ See, for example, H. Kopferman, *Nuclear Moments* (Academic Press Inc., New York, 1958), 2nd ed., pp. 150-152.

⁷ G. Araki, Proc. Phys. Math. Soc. Japan **19**, 128, 592 (1937).

⁸ H. Bucka and H. J. Schussler, Ann. Physik **7**, 225 (1961).

In Table II are given present and previously measured quantities relating to the lifetime of the 1P_1 and 3P_1 state of Ba. Our results are in excellent agreement with the recent measurements of Hulpke, Paul, and Paul. It is apparent from Fig. 1 that one cannot determine the oscillator strength for the 5535-Å or 7911-Å lines from a knowledge of the lifetime of the 3P_1 and 1P_1 states alone since transitions can also occur from these states to the $6s5d\ ^3D_{3,2,1}$ and 1D_2 levels which lie lower than the $6s6p\ ^3P_{2,1,0}$ and 1P_1 levels, respectively. One may, however, determine the intermediate coupling coefficients c_1 and c_2 by the derivation of the fine structure from an interval rule (Wolfe method) and also by the ratio of the oscillator strengths which has been measured by Prokofjew (see Table II). These two methods yield almost identical results and are listed in Table III.

From the results listed in Table II, one can determine the oscillator strength of the 27 750 and 29 222 lines as follows: the lifetime of the 1P_1 state is almost completely

TABLE II. Experimental and theoretical quantities relating to the lifetime of the 1P_1 and 3P_1 state of Ba.

Author	Quantity determined
a. Experimental	
Prokofjew ^a	$f(5535)/f(7911) = 146 \pm 2$
Wessel ^b	$f(5535) = 1.8 \pm 0.25$
Ostrovskii, Penkin, Shabanova ^c	$f(5535) = 1.7 \pm 0.2$
Bucha, Schüssler ^d	$\tau(^1P_1) = (10.4 \pm 0.5) \times 10^{-9}$ sec
Penkin, Shabanova ^e	$f(5535) = 1.40 \pm 0.05$
Hulpke, Paul, Paul ^f	$\tau(^1P_1) = 8.36 \pm 0.25 \times 10^{-9}$ sec
Present work	$\tau(^1P_1) = (8.20 \pm 0.20) \times 10^{-9}$ sec
Bucha, Nagel ^g	$\tau(^3P_1) = (1.21 \pm 0.12) \times 10^{-6}$ sec
b. Theoretical	
Bates and Damgaard ^h	$f(5535) = 1.92$

^a W. Prokofjew, Z. Physik **50**, 701 (1928).

^b G. Wessel, Z. Physik **126**, 440 (1949); also **130**, 106 (1951).

^c Yu. I. Ostrovskii, N. P. Penkin, and L. N. Shabanova, Dokl. Akad. Nauk SSSR **120**, 66 (1958) [English transl.: Soviet Phys.—Doklady **3**, 538 (1958)].

^d H. Bucka and H. J. Schussler, Ann. Physik **7**, 225 (1961).

^e N. P. Penkin and L. N. Shabanova, Opt. i Spektroskopiya **12**, 3 (1962) [English transl.: Opt. Spectry. (USSR) **12**, 1 (1962)].

^f E. Hulpke, E. Paul, and W. Paul, Z. Physik **177**, 257 (1964).

^g H. Bucka and H. H. Nagel, Ann. Physik **8**, 329 (1961).

^h See Ref. 4, Table I.

TABLE III. Intermediate coupling coefficients.

Method of Determination	Magnesium		Barium	
	c_1	c_2	c_1	c_2
a	0.5757	0.8177	0.5000	0.8650
b	0.5742	0.8187	0.4938	0.8695
Pure L - S coupling	0.5774	0.8165	0.5774	0.8165

^a From ratio of oscillator strengths $f(^3P_1 \rightarrow ^1S_0)/f(^1P_1 \rightarrow ^1S_0)$.
^b From deviation of 3P state fine structure from an interval rule.

determined by its decay to the 1S_0 state since this is an allowed transition. Also the oscillator strength of the $\lambda(^1P_1 - ^1D_2) = 15\,000 \text{ \AA}$ line has been estimated by Hulpke, Paul, and Paul. Averaging their $\tau(^1P_1)$ result and ours we have $f(5535) = 1.55 \pm 0.06$, where we have taken $f(15\,000) = 0.17 \pm 0.07$. From the Prokofjew ratio given in Table II, we obtain $f(7911) = (1.06 \pm 0.05) \times 10^{-2}$. The lifetime of the 3P_1 state may be written in terms of the spontaneous transition probabilities as $1/\tau(^3P_1) = A(7911) + A(27750) + A(29222) = 4.14 \times 10^6 + 3.59A(27750)$ where we have used the theoretical result $A(27750) = 0.386A(29222)$. We find from this equation that $A(27750) = 1.18 \times 10^{-5} \text{ sec}^{-1}$ or $f(29222) = 0.0136 \pm 0.0015$ and $f(27750) = 0.0130 \pm 0.0015$.

HYPERFINE STRUCTURE OF Ba^{137}

The energy level diagram of Ba^{137} ($I = \frac{3}{2}$) is shown in Fig. 2 and has been calculated using the values of the dipole coupling constant A and the quadrupole coupling constant B given by Jackson and Tuan.⁹ These values are $A = -108.3 \pm 3.0 \text{ Mc/sec}$ and $B = 58.2 \pm 3.0 \text{ Mc/sec}$. The zero-field hfs splittings are given by the expressions

$$\Delta\nu(\frac{5}{2}, \frac{3}{2}) = 5A/2 + 5B/4$$

and

$$\Delta\nu(\frac{3}{2}, \frac{1}{2}) = 3A/2 - 9B/4.$$

The circled intersection in Fig. 2 of the $F = \frac{5}{2}$ $m = \frac{5}{2}$ and the $F = \frac{3}{2}$ $m = \frac{1}{2}$ levels is the only observable level crossing for the 1P_1 state of Ba^{137} . The location of this crossing is given by the expression⁵

$$g_J \mu_0 H = 2A(1 - B/4A)(1 + B/2A)/(1 + B/4A). \quad (1)$$

This expression for the field at which the level crossing occurs is quite insensitive to B/A . This can be seen as follows: if one takes Jackson and Tuan's values for A and B , then the maximum uncertainty in this is $B/A = -0.537 \pm 7\%$ or $-0.575 \leq B/A \leq -0.499$. Using these two extreme values of B/A we find from Eq. (1) that $g_J \mu_0 H = 2A(0.964)$ for $B/A = -0.499$ and $g_J \mu_0 H = 2A(0.952)$ for $B/A = -0.575$ so that we have $g_J \mu_0 H = 2A(0.958 \pm 0.006)$.

The experimental arrangement was the same as in the lifetime experiments except that a 38-cps modulat-

⁹ D. A. Jackson and D. H. Tuan, Phys. Rev. Letters **11**, 209 (1963).

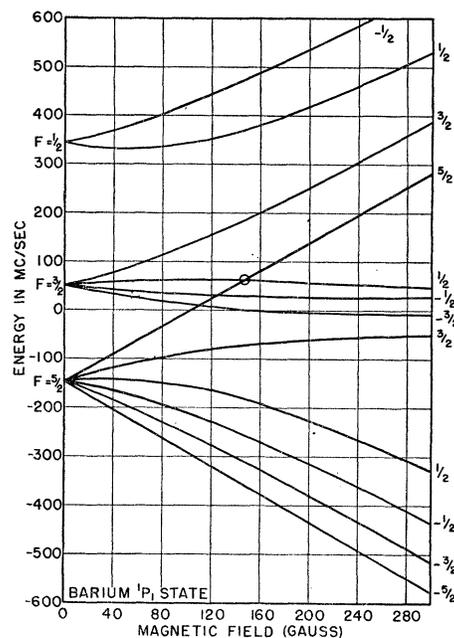


FIG. 2. Magnetic field dependence of the Ba $6s6p \ ^1P_1$ state hfs. The circled intersection of levels is the one observable high-field level crossing.

ing field was applied to the atoms along the static magnetic field direction so that lock-in techniques could be used. Also the atomic beam was produced from a sample enriched to 43.6% in Ba^{137} .

The magnetic field at which the level crossing occurred was $151.3 \pm 0.7 \text{ G}$ and had a full half-width when extrapolated to zero modulation field of about 27.5 G, in good agreement with the expected linewidth. When one takes $g_J = 1.025$ we find that $A = -113.2 \pm 1.0 \text{ Mc/sec}$ which is in good agreement with the value of Jackson and Tuan. Since only one observable level crossing occurs in the 1P_1 state of Ba^{137} we cannot obtain B from this crossing, as indeed we could not obtain A either if it were not for Jackson and Tuan's approximate value for B .

$A(^1P_1)$ may be estimated theoretically for the Ba $6s6p$ configuration provided $A(^3P_2)$ and $A(^3P_1)$ are known with sufficient precision. The presently known¹⁰ value of $A(^3P_2)$ (obtained from optical spectroscopy) is not sufficiently precise to make a meaningful estimate of $A(^1P_1)$. We may, however, compare Q as obtained from Jackson and Tuan's value for B in the 1P_1 state with that obtained by Zu Putlitz¹¹ from B in the 3P_1 state. We have

$$Q = \frac{15\mu_B^2 Z_i H_r(e=1; Z_i) B}{e^2 [c_2^2 R_r^1(Z_i) + 2\sqrt{2} c_1 c_2 S_r(Z_i)] \delta \bar{W}}, \quad (2)$$

¹⁰ Landolt-Börnstein Tables (Springer-Verlag, Berlin, 1952), Vol. I, part 5.

¹¹ Von G. Zu Putlitz, Ann. Physik **7**, 248 (1963).

where the notation is the same as given by Zu Putlitz. From (2) we obtain $Q(137) = 0.20 \times 10^{-24}$ cm² which does not agree too well with Zu Putlitz's result of $Q(137) = 0.28(3) \times 10^{-24}$ cm². The disagreement is outside experimental error, and may be due to the 1P_1 and 3P_1 states having different values of $\langle 1/r^3 \rangle_{\text{av}}$. From experience with other members of the group II elements,

however, we would expect the result obtained from the 3P_1 state to be more reliable.

ACKNOWLEDGMENTS

The author wishes to thank Dr. A. Gallagher for several helpful discussions and to acknowledge the help of R. Holohan in all phases of the experimental work.

Cross Sections for Ion and Electron Production in Gases by Fast Helium Ions (0.133–1.0 MeV). I. Experimental*

R. A. LANGLEY,† D. W. MARTIN, D. S. HARMER, J. W. HOOPER, AND E. W. MCDANIEL

Georgia Institute of Technology, Atlanta, Georgia

(Received 11 May 1964)

This paper reports the measurement of apparent cross sections for production of slow positive ions and free electrons by He⁺ ions incident on He, Ne, Ar, H₂, N₂, O₂, and CO in the energy range 0.133 to 1.0 MeV, and similar cross sections for He⁺⁺ ions incident on He and H₂ in the energy range 0.5 to 1.0 MeV. Direct comparisons are made with experimental results obtained by other investigators for He⁺ ions at energies up to 0.40 MeV. In the companion paper comparisons are also made with available theoretical calculations and with results predicted from the Bethe-Born approximation with the evaluation of certain parameters from earlier measurements with proton projectiles.

I. INTRODUCTION

WE have measured the apparent cross sections for production of slow positive ions and free electrons by He⁺ ions incident on helium, neon, argon, hydrogen, nitrogen, oxygen, and carbon monoxide for projectile energies over the range 0.133 to 1.00 MeV. We have also measured these cross sections for He⁺⁺ ions on hydrogen and helium over the energy range 0.50 to 1.00 MeV. Measurements of these quantities by other investigators have been confined to lower energies.

The experiments described here constitute a segment of a continuing program of absolute determinations of the cross sections for ion and electron production at high energies by various light ions and atoms on the same targets. The results for protons incident on the target gases listed above have already been reported.^{1,2} A detailed comparison of the proton results with the available theoretical calculations and with data for incident electrons has also been published.³ Similar

comparisons of the present results with theory and with the proton data are presented in the following paper (II).

We selected H₂ and He as target gases because of their importance in controlled fusion research and because their structural simplicity has permitted theoretical calculations of their cross sections. Calculations have not yet been made on the other noble gases, but Ne and Ar were studied because of the general interest in these gases and because of the desirability of obtaining data on heavier atomic systems. Also included are N₂ and O₂ since they are atmospheric constituents of considerable practical interest. Finally CO was included because it is a common contaminant in vacuum systems and because the comparison of data on CO and N₂ is of interest. These two molecules have the same number of electrons and somewhat similar structures.

The present results were presented at the Third International Conference on the Physics of Electronic and Atomic Collisions in London (July 1963) and a summary will be published in the Proceedings (North-Holland Publishing Company, Amsterdam, 1964). However, limitations on the permitted text length necessitated the omission of important details which are given here. Also, comparison is made here with results recently published by other workers.

II. EXPERIMENTAL METHODS

In all our high-energy collision studies, the projectile source has been a 1-MeV Van de Graaff positive-ion

* This work was partially supported by the Controlled Thermonuclear Research Program of the U. S. Atomic Energy Commission.

† Present address: CRUZ, Air Force Cambridge Research Laboratory, Bedford, Massachusetts. Portions of the results presented here were included in a thesis submitted by RAL to the faculty of the Georgia Institute of Technology in partial fulfillment of the requirements for the degree of Doctor of Philosophy.

¹ J. W. Hooper, E. W. McDaniel, D. W. Martin, and D. S. Harmer, *Phys. Rev.* **121**, 1123 (1961).

² E. W. McDaniel, J. W. Hooper, D. W. Martin, and D. S. Harmer, *Proceedings of the Fifth International Conference on Ionization Phenomena in Gases (Munich, 1961)* (North-Holland Publishing Company, Amsterdam, 1962), Vol. I, p. 60.

³ J. W. Hooper, D. S. Harmer, D. W. Martin, and E. W. McDaniel, *Phys. Rev.* **125**, 2000 (1962).