Mean lives for the $5p\,3/2[1/2]_1$ and $5p\,1/2[1/2]_1$ levels in singly ionized rubidium (Rb II)†

M. Gaillard
Laboratoire de Spectrométrie Ionique et Moléculaire, Université de Lyon, 43 Boulevard du 11 Novembre 1918, 69100 Villeurbanne, France

H. J. Andrä, A. Gaupp, W. Wittmann, and H.-J. Plöhn
Institut für Atom- und Festkörperphysik (A), Freie Universität Berlin, 1000 Berlin 33, Boltzmannstrasse 20, Germany

John O. Stoner, Jr.
Department of Physics, The University of Arizona, Tucson, Arizona 85721
(Received 14 April 1975)

We have measured the mean lives of two levels in Rb II by exciting them selectively in fast beams of metastable Rb^+ ions with radiation from an argon-ion laser, and observing their time-resolved decay downstream. The results are for $5p\,3/2[1/2]_1$, 8.04 ± 0.08 nsec, and for $5p\,1/2[1/2]_1$, 6.9 ± 0.2 nsec. We describe some of the systematic errors that must be minimized in order that the accuracies obtained by this method approach or surpass those obtained by other means. Our results show clearly that advanced calculational methods are necessary in order to take full advantage of the experimental accuracy.

I. INTRODUCTION

We have applied the method of selective excitation of fast ionic beams by light from an argon-ion laser to the study of the mean lives of two levels in Rb II. The principles of this method have been described previously.[1-3] In this method, a laser beam crosses a beam of fast ions at an angle such that a Doppler-shifted spectral line characteristic of the ions matches a wavelength emitted by the laser. We monitor the light emitted in a decay transition involving the upper level as a function of distance downstream from the interaction region, and in this way measure the mean life of the upper level.

Because contributions to the observed signals due to cascades into the upper level from still higher levels are largely removed by this technique, it appears possible to measure mean lives of atomic and ionic levels to better than 1%. We indicate here the nature of some of the precautions that must be followed in order to accomplish a measurement at that level.

This work substantially improves our knowledge of the mean lives of the $5p\,\frac{3}{2}[\frac{1}{2}]_1$ and $5p\,\frac{1}{2}[\frac{1}{2}]_1$ levels in Rb II. This permits the direct test of two semiempirical methods for calculating mean lives of ions in the noble-gas isoelectronic sequences. We find that these methods are inadequate to explain our results. It appears that such calculations must be modified to include the effects of large numbers of configurations if they are to approach the accuracy now available in experiments.

II. EXPERIMENTAL METHOD

A. Ionic beam

We directed singly-charged positive ions from a Danfysik 350-kV accelerator through a switching magnet and thence to an aluminum target chamber. Isotopic separation was done magnetically at the high-voltage end of the accelerator; this helped ensure that the ionic beam passed by the analyzing magnet was isotopically pure. In any case, the excitation method is strongly selective toward only one level in one isotope at a time, except when the laser and ionic beams are nearly perpendicular. Electromechanical scanners monitored profiles at several positions along the ionic beam to verify that no unwanted beams were present and that the desired beam approached the chamber properly. Most of our data were taken with the mass-85 isotope, since it is the more abundant.

Immediately before entering the chamber, the ions passed through a cryogenically pumped gas cell. (Collisions by the ions with gas atoms in this cell served to excite some of the ions to metastable levels which could later be further excited by light from the laser.) The ions then entered the chamber through a rectangular aperture 3.5 mm wide and 3.0 mm high, and passed through the chamber to a set of defining apertures. Only those ions passing through a 5-mm-diameter hole in a grounded plate 450 mm from the entrance aperture were monitored. Those that passed through a slot 3.0 mm wide centrally located behind that hole were collected on a stainless-steel monitor plate; the current from this plate indicated the intensity of the ion beam. Currents collected by copper jaws on each side of the central slot indicated whether the ion beam was approximately located on its centerline. Total currents passing through the circular aperture ranged from 6 to 25 μA, with typically 10–20% on each jaw.

The gas cell consisted of a chamber having a length of 85 mm, with apertures 5 mm in diameter at each end. Butane (C_4H_{10}) vapor was admitted

into this chamber to pressures of $\sim 10^{-3}$ Torr. Vapor effusing from the apertures was trapped on surfaces held at $-190\,^{\circ}$C by a reservoir of liquid nitrogen. During operation of the cell, pressures in the remainder of the vacuum system ranged from 7×10^{-6} to 2×10^{-5} Torr.

Small magnetic fields ($\simeq 1$ G) in the chamber would have caused Zeeman splittings of the order of 1 MHz in the upper levels studied here. The upper states were excited coherently, since the time required for the ion to pass through the interaction region was only of the order of a nanosecond; thus, the conditions for the excitation of quantum beats were satisfied. Such beats would superimpose periodic oscillations on the decay curve.[4] The fractional error in a mean life τ introduced by the presence of such beats is of the order of $\omega_L\tau$, where ω_L is the Larmor frequency. We reduced the earth's magnetic field on the axis of the ionic beam to less than 50 mG (using coils external to the chamber) so that $\omega_L\tau \lesssim 0.001$ due to this.

B. Energy measurements

We measured the particles' energies several times during each run by removing the monitor plate and allowing the ions to enter an electrostatic analyzer. This analyzer was calibrated using quantum beats in helium[5] and checked occasionally by careful measurement of the angle between ionic beam and laser beam at which resonance occurred. We used tabulated ionic masses[6] to transfer energy measurements into velocity measurements.

In no case did the apparent energy vary by as much as 1 keV during a run; this implies that the particles' speeds were constant to within about 0.16% at 310 keV and 0.28% at 180 keV. The ions presumably lost kinetic energy and increased their spread in velocity as they passed through the gas cell. However, we observed no effects that could be attributed to an energy spread.

C. Illumination of the ionic beam

Most of the data reported here were taken with the use of a Spectra-Physics Model 170 argon-ion laser operating continuously on one line at a time in many longitudinal modes. The spectral width of the laser "line" at λ 4726.859 Å was measured with a Spectra-Physics Model 470 spectrum analyzer, and found to be about 2 GHz at minimum power ($\simeq 50$ mW), rising to about 4 GHz at maximum power ($\simeq 2.5$ W). The laser could also be operated "single-mode" by introducing an étalon into the laser cavity, in which case it oscillated on two or three longitudinal modes with spacings about 70 MHz.

Some preliminary data were taken with the use of a Spectra-Physics Model 164 laser which provided only 240 mW (multimode operation) at λ 4726.859 Å.

The beam from the laser was directed into the chamber and across the ion beam via several front-surface mirrors, so that the laser and ionic beams lay in a nearly horizontal plane (Fig. 1). The multiple reflections depolarized the laser beam so that its residual linear polarization was less than 10% at the ionic beam.

The mirror closest to the ion beam was mounted on a vertical shaft which could be rotated by a reversible dc motor operating through a double worm-gear drive. The resistance of a ten-turn potentiometer mounted on one worm indicated the orientation of the mirror and hence the angle θ between the laser and ion beams. This angle could be measured geometrically to within about $\pm 0.2^{\circ}$. It was set before each run as closely as possible to the angle implied by the formula for the wavelength (λ_{laser}) absorbed by a particle absorbing wavelength λ_{ion} in its rest frame and moving with speed v:

$$\lambda_{\text{laser}} = \lambda_{\text{ion}} \frac{[1-(v/c)\cos\theta]}{[1-(v^2/c^2)]^{1/2}} .$$

Without any lenses in the path of the laser beam, its diameter was about 4 mm at the ionic beam. In order to reduce the width of the interaction region, we could place either a cylindrical or spherical lens with focal length 300 mm in the laser beam about 300 mm from the ionic beam. In such cases the observed width of the interaction region was about 0.5 mm.

After the laser beam crossed the ionic beam, it was absorbed in a metal dump or reflected by an auxiliary mirror into a distant corner of the vacuum system. We placed black nonmagnetic baffles at strategic points within the chamber to reduce the stray laser light to acceptable levels.

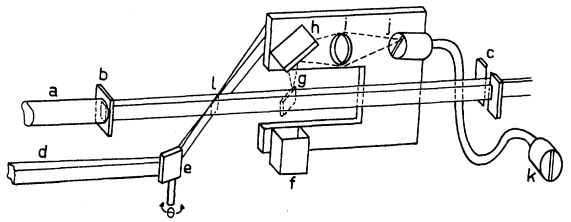

FIG. 1. Schematic diagram of the excitation and detection regions: (a) ionic beam entering from left; (b) entrance aperture; (c) differential aperture for centering the ionic beam; (d) laser beam entering from left; (e) tuning mirror; (f) light shield; (g) detection region; (h) mirror; (i) lens; (j) entrance end of light guide; (k) exit end of light guide; (l) excitation region.

The angular divergence in a vertical plane of the part of the ionic beam excited by the laser was expected to be essentially independent of the method of illumination and to have no significant effect upon the optical collection efficiency of the detection optics.

The angular divergence in the horizontal plane of the part of the ion beam excited by the laser was expected to depend upon whether the laser was operated with the étalon ("single-mode"), whether a lens was used, and upon the laser power (which determined to some extent its linewidth). If the collection efficiency for the detection optics were to depend significantly upon the width or position of the excited ion beam in the horizontal plane, we would expect to see significant variations in apparent mean lives as the angular divergence of the excited beam was changed. We attempted to minimize the probable effects of horizontal divergence of the ion beam by appropriate transverse location of the collection optics (see next section). We also searched for systematic changes of the apparent mean life of the $5p\,\frac{3}{2}[\frac{1}{2}]_1$ level with the method of illumination of the ion beam.

D. Detecting optics

Part of the light emitted by the ionic beam was collected by a lens-mirror combination above the beam; the beam was imaged at unity magnification onto one end of a flexible quartz fiber array (Fig. 1). That array had lateral diminsions 1×20 mm, masked at the entrance end to 1×9 mm. The entrance end was oriented about the optical axis so that the image of the array formed by the lens crossed the ionic beam at about the same angle as did the laser beam. Below that image we placed a blackened cavity which served to reduce the amount of light scattered into the optics. The lens, mirror, cavity, and entrance end of the fiber array were mounted on a carriage which could be moved parallel to the axis of the ionic beam by a precision screw driven by a stepping motor.

We attempted to measure the relative collection efficiency of the detection optics as a function of downstream position of the carriage. By monitoring the signal from a small lamp fixed to the carriage, we ascertained that flexure of the fiber optic caused no changes larger than 0.1% in collection efficiency. We used the light emitted by the ionic beam excited by residual gas and light emitted by the residual gas itself, with the laser beam blocked and the input valve to the gas cell closed, to simulate the laser-excited beam. In this way we determined that the overall collection efficiency of the detection optics decreased nearly linearly by about 1% over 50 mm travel of the carriage. This measurement was supported by observations of the background present during measurements on the $5p\,\frac{1}{2}[\frac{1}{2}]_1$ level, which was observed to be constant within $\pm 1\%$ along the part of the ionic beam used. It was also supported by observations on the constancy of the signal vs downstream position of the carriage during the optimization of the lateral position of the carriage (see below), which placed a limit of $\pm 2\%$ on any changes in collection efficiency.

The optimum lateral position of the collecting optics was found by maximizing the signal due to light scattered by dust in the air into the fiber array, when a laser beam was passed along the axis of the ionic beam. We determined in this way that the optical collection efficiency for the optics did not vary by more than about 2% for lateral translations of ± 1 mm from the best position.

The maximum length of the ionic beam used for determination of lifetimes was about 50 mm, corresponding to about four decay lengths. The effects of any divergence of the excited ionic beam (maximum about 0.012 rad set by properties of the laser beam and somewhat less as set by the physical collimation of the ionic beam), coupled with changes in the lateral sensitivity of the collecting optics, were therefore expected to introduce systematic errors into measured lifetimes at the level of 0.1% or less.

The exit end of the fiber array had dimensions 1×20 mm, and served as the entrance slit of a small grating spectrometer. With the width of the exit slit set at 1.3 mm, this instrument had a resolving limit full width at half-maximum (FWHM) of 16 Å. A relay lens focused light leaving the exit slit onto the photocathode of a cooled EMI-9659Q photomultiplier operated in a pulse-counting mode.

With this system, the background signal due to scattered laser light was nearly negligible when the spectrometer was set at the lines used for detection.

E. Electronics

The longitudinal position of the carriage was controlled via a Digital Equipment Corp. PDP 8/e Minicomputer which drove the stepping motor. During a run, light was collected at equally spaced intervals along the axis of the ionic beam for times normalized to the charge collected on the monitor plate. Pulses from the photomultiplier passed through an amplifier and discriminator, then were counted in channels (within the computer) which were changed synchronously with the position of the carriage.

F. Data acquisition

After adjusting the pressure in the gas cell and the direction of the laser beam to maximize the desired signal, we studied the structure of each absorption line by scanning the angle θ through the range for which the resonance occurred. In cases where hyperfine structure was evident, we then set the value of θ such that the strongest component(s) were excited.

We obtained a "lifetime curve" by programming the computer to acquire data at a sequence of equally spaced points along the axis of the ion beam. These points usually included the excitation region and about five decay lengths downstream. Typically, data were taken at 128 points spaced at 0.5 mm; such a scan required about 2 min, and 10 or 20 such scans were co-added in the computer to form one set of data. The total counting time was recorded for each set. By adding many scans, each of short duration, we reduced systematic changes in the apparent lifetime due to slow drifts of the laser power, focus and/or wavelength, pressure in the gas cell, background pressure, or dark count rate. Occasionally, scans to determine background were made with the laser and/or the ion beam blocked.

G. Analysis

To extract a lifetime from a set of data, we first subtracted normalized backgrounds due to scattered laser light, excitation of the ion beam by residual gas, and dark count of the photomultiplier. The data points from the region well past where excitation occurred were then weighted by their standard deviations and least squares fitted by a single exponential plus an adjustable constant. In most cases the data were fitted well by this procedure, as indicated by chi-squared (χ^2) tests. In the cases where the data were poorly fitted, they were discarded. (In no case did we find evidence for any real systematic departure from exponential decay.)

By plotting χ^2 vs constant assumed background level, we could identify the sensitivity of χ^2 and mean life to background level. In this way we determined that a mean life for a data set was defined easily within 0.5% in cases for which the peak count rate was several thousand counts per channel. The spread of mean-life values in a sequence of nominally identical measurements was observed to be typically 2% in such cases.

III. EXPERIMENTAL RESULTS

A. The $5p\ ^3_2[^1_2]_1$ level

The Rb^+ ions emerging from the accelerator were presumably almost entirely in the ground level $3d^{10}4s^24p^6\,^1S_0$. We used the population of ions in the metastable level $5s\ ^3_2[^3_2]^o_2$ (energy 133 341.37 cm^{-1}, 16.25 eV) produced by collisions in the gas cell and with residual gas, and induced the transition $5s\ ^3_2[^3_2]^o_2 - 5p\ ^3_2[^1_2]_1$ by the Ar^+ laser line at λ 4764.862 Å.[7] The laser line was Doppler-tuned to the ionic transition at λ 4775.949 Å[8] by causing the two beams to intersect at $\theta = 33.8° \pm 0.2°$ with Rb-85 ions accelerated to energies of about 309.6 keV, or Rb-87 ions accelerated to about 316.9 keV. Excitation of the upper level was detected by observing light from the transition $5s\ ^3_2[^3_2]^o_1 - 5p\ ^3_2[^1_2]_1$ at λ 5152.115 Å.

The tuning curve (dependence of signal upon θ, with position of the carriage fixed) showed structure arising from hyperfine splitting in the $5s\ ^3_2[^3_2]^o_2$ level for both Rb-85 and Rb-87 (Fig. 2). These structures have been studied in detail.[9] For the purpose of estimating an upper limit to the angular divergence of the accelerator beam, we assumed that the strongest peak was a single component of the hyperfine multiplet. With the laser operated in "single-mode," and with no lens in the laser beam, the width (FWHM) of the resonance corresponded to a tilt of the laser beam of about 12 mrad. This provided then an upper limit to the effective divergence of the ion beam in the horizontal plane.

The hyperfine splittings in the upper levels studied here were all of the order of 1 GHz or greater.[9] Our optical detection system collected light from a segment of the ionic beam at least 1 mm in length, corresponding to a range of detection times greater than 1 nsec. The excitation region itself had a width along the axis of the ionic beam of about 0.5 mm, corresponding to a range of excitation times greater than 0.5 nsec. On these bases we expected no beats corresponding to upper-level hfs splittings to appear on our decay

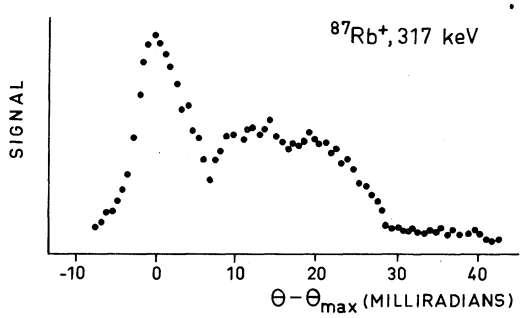

FIG. 2. Tuning curve for excitation of the $5p\ ^3_2[^1_2]_1$ level in ^{87}Rb II, showing unresolved hyperfine structure in the lower level. For lifetime measurements, the angle θ between the laser beam and the ionic beam was set at θ_{max} to yield maximum signal.

curves, and in fact no indications of beats were seen.

Nineteen useful sets of data on the $5p\,^3\!/_2[1/2]_1$ level were obtained with slightly different apparatus on three different days. These yielded lifetimes ranging from 7.85 to 8.23 nsec; a histogram showing the distribution of values for the lifetime of this level is shown in Fig. 3.

No *statistically significant* differences in apparent mean life could be identified with laser power, method of illumination or laser bandwidth. Although some indications of a dependence of the apparent mean life upon isotope or angular offset of the ionic beam could be seen (Fig. 3), we do not find them convincing. Our final value (Table I) for the lifetime of the $5p\,^3\!/_2[1/2]_1$ level is the mean of all measurements, excluding those taken with an angular offset or with the mass-87 isotope. A correction has been made for a 1% decrease in optical collection per 50 mm carriage displacement downstream. The systematic errors expected to contribute to this measurement are shown in Table II. The standard deviation of the mean of 15 measurements having an internal standard deviation of 0.06 nsec is 0.02 nsec, assuming a normal (Gaussian) distribution. We suggest that a conservative error limit for our combined measurements for the lifetime of this level is ± 0.08 nsec, and our final value for the lifetime of this level is then 8.04 ± 0.08 nsec.

B. The $5p\,^1\!/_2[1/2]_1$ level

We excited this level from the metastable $4d\,^3P_2^o$ level (energy 143 955.66 cm^{-1}, 17.9 eV) which was produced by collisions in the gas cell and residual gas. The ionic transition for excitation occurs at λ4730.454 Å;[8] this was Doppler tuned to the Ar^+ laser line at λ4726.859 Å[7] by causing the two beams to intersect at about $\theta = 70.2°$ with Rb^+-85 ions at a nominal energy of 180 keV. We detected the upper level by the appearance of the line at λ4530.337 Å due to the transition

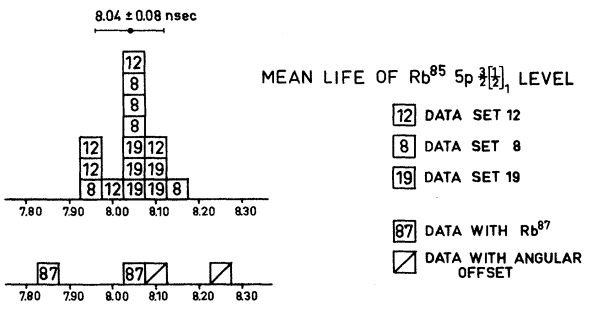

FIG. 3. Histogram showing the data from which the mean life of the $5p\,^3\!/_2[1/2]_1$ level in Rb II was derived.

TABLE I. Mean lives of levels in Rb II.

	This work	Kumar *et al.* (Ref. 9)	Theory
$5p\,^3\!/_2[1/2]_1$	8.04 ± 0.08 nsec	8.3 ± 1.2 nsec	14 nsec [a] 10 nsec [b]
$5p\,^1\!/_2[1/2]_1$	6.9 ± 0.2 nsec	•••	3.7 nsec [a] 3.2 nsec [b]

[a] Calculated using the methods of Refs. 11 and 12.
[b] Calculated using the methods of Ref. 15.

$4d\,^3P_0^o - 5p\,^1\!/_2[1/2]_1$. Neither the tuning curve nor the decay showed indications of hyperfine structure.

Table I shows the mean of six different data sets taken on one day. Counting rates for these observations were noticeably lower, and the background due to cascades from higher levels excited by collisions in the gas cell and residual gas was higher than in the runs to determine the lifetime of the $5p\,^3\!/_2[1/2]_1$ level. We have therefore extended the uncertainty in this mean life to include all of the individual experimental values. The decay lengths for these data were less than for the $5p\,^3\!/_2[1/2]_1$ data because of the smaller lifetime and because these measurements were made at lower energy. Thus, all of the systematic errors discussed above for the lifetime of the $5p\,^3\!/_2[1/2]_1$ level should be less for the $5p\,^1\!/_2[1/2]_1$ level. Our final experimental value for the mean life of this level is 6.9 ± 0.2 nsec.

IV. DISCUSSION

A. Systematic errors

The principal difficulties associated with making an accurate measurement of an atomic mean life using a time-of-flight method appear to be in knowing the velocities of the emitting particles and in ensuring that the optical collection efficiency is either constant or varies in a known manner with position. Deviations from constancy can occur due to changes in the optical components, the ion beam, or their relative position. Changes in the optics were avoided by mounting them rigidly to a common plate and by avoiding strong curvatures of the fiber array as the carriage moved. The

TABLE II. Systematic errors in the determination of the mean life of the $5p\,^3\!/_2[1/2]_1$ level in Rb II.

Velocity uncertainty		± 0.03 nsec
Magnetic fields	less than	± 0.01 nsec
Optical collection efficiency		± 0.01 nsec
Angular offset of the ion beam	less than	± 0.02 nsec
Total of absolute values	less than	0.07 nsec
Square root of the sum of the squares of absolute values		± 0.04 nsec

nominal axis of the ion beam was parallel to the motion of the carriage to better than 0.001 rad. The moving parts within the chamber were located sufficiently far from the ion beam that no part of the excited beam could be intercepted by them. Finally, the angular divergence and offset of the excited beam, as measured by the differential aperture and by the tuning curve, were kept small enough that they were not expected to contribute significantly to systematic error in measurement.

As discussed previously (Sec. IID), we had some evidence for a linear decrease in sensitivity with position downstream for the collecting optics. A decrease by a small fractional part f over length L changes apparent mean lives by the factor $(1-fX_0/L)$, where X_0 is the true decay length for the excited ions. The quantity $+(fX_0/L)\tau$ is 0.01 nsec for the transitions studied here; this correction has been included in all our experimental values quoted in this paper. Because there still remains the question of whether our simulations of the laser-excited beam were completely accurate, we have included an uncertainty of ± 0.01 nsec in Table II due to uncertainties in collection efficiency.

We assume that our methods of measurement and analysis introduce no systematic errors into a mean life obtained from an ensemble of measurements, since the errors in the "true" background due to statistical fluctuations in the backgrounds should be distributed symmetrically about the mean value.

B. Other measurements

Kumar *et al.* have measured the mean lives of the $5p\,{}^3_2[\frac{1}{2}]_1$ level by methods of beam-foil spectroscopy (BFS).[10] Their result is included in Table II. Our results do not conflict with theirs. We note that in the present work the almost complete absence of cascades into the upper levels permits assignment of error limits smaller by an order of magnitude than in BFS.

C. Theoretical estimates

We are aware of no published theoretical studies of the lifetimes of the two levels of interest. On the other hand, several methods have been developed recently for the calculation of transition probabilities in the rare gases,[11] and attempts have been made[12] to extend these techniques to isoelectronic ions.

In order to get some theoretical estimates for comparison with our new experimental results, we used the method of Aymar *et al.*,[11] in which a first approximation to the lifetimes of the np levels of noble gases can be obtained without any intermediate-coupling calculations using the formula

$$\frac{1}{\tau}(\sec^{-1}) = 2.026\times 10^{18}\sum (nl\,|r|\,n'l')^2(2l'+1) \times \begin{Bmatrix} l & 1 & l' \\ 0 & 0 & 0 \end{Bmatrix}^2 \frac{1}{\bar{\lambda}^3_{n'l'}(\text{Å})^3}.$$

Here $(nl\,|r|\,n'l')$ is the reduced matrix element for the transition, in atomic units. This assumes that one may define the average wavelengths $\bar{\lambda}_{n'l'}$ for the $n'\,s$-np and $n'\,d$-np decay transitions using the relationship

$$\frac{1}{\bar{\lambda}^3_{n'l'}} = \frac{1}{p}\sum_k \frac{1}{\lambda^3_k}$$

where p is the number of allowed transitions and λ_k is the wavelength of a transition in the multiplet $n'l'-np$. At this level of approximation for the lifetime τ, the Coulomb approximation[13,14] provides a sufficiently accurate value for the integral $(nl\,|r|\,n'l')$.

Table I shows the results of this calculation. The failure of this method to give better than an order-of-magnitude estimate may be traced to the fact that in Rb II, the splitting of the $5p$ configuration is comparable to the $5s$-$5p$ and $4d$-$5p$ intervals. This makes the definition of suitable average wavelenghts rather arbitrary.

As an alternative we used the semiempirical intermediate-coupling method described by Statz *et al.*,[15] and the tables of intermediate-coupling coefficients listed by Reader and Epstein[8] for the levels in Rb II. In this method the spontaneous transition probability between an initial level

$$|\psi\rangle = \sum_i \alpha_i |c_i L_i S_i J M\rangle$$

and a final level

$$|\psi\rangle = \sum_j \beta_j |c_j L'_j S'_j J' M'\rangle$$

is given by

$$A(J,J')(s^{-1}) = \frac{2.026\times 10^{18}}{(2J+1)\lambda(\text{Å})^3}\left|\sum_{ij}\alpha^*_i\beta_j\left\langle c_i L_i S_i J\left\|\sum r_m C^{(1)}_m\right\| c_j L'_j S'_j J'\right\rangle\right|^2.$$

The matrix elements of $r_k C^{(1)}_k$ [the product of the position operator for the kth electron and the spherical Racah tensor of rank 1] are given for transitions of the sort [(core)nl–(core)$n'l'$] by

$$\left\langle nlLSJ \left\| \sum r_m C_m^{(1)} \right\| n'l'L'S'J' \right\rangle = \delta_{SS'}(-1)^{J'+S+l'+l+\bar{L}}[(2J+1)(2J'+1)(2L+1)(2L'+1)(2l+1)(2l'+1)]^{1/2}$$
$$\times \begin{Bmatrix} S & L & J \\ 1 & J' & L' \end{Bmatrix} \begin{Bmatrix} \bar{L} & l & L \\ 1 & L' & l' \end{Bmatrix} \begin{Bmatrix} l & 1 & l' \\ 0 & 0 & 0 \end{Bmatrix} (nl|r|n'l')$$

where $\bar{L}$ is the orbital angular momentum of the core.

Again we used the Coulomb approximation to evaluate the radial integrals (the same for both upper levels). The resulting lifetimes are shown in Table I. Despite its increase in complexity, this second calculation yielded no marked improvement over the first. Since the wave functions of Ref. 8 include mixing of the 5*s* and 4*d* configurations, our calculated transition probabilities could not be separated into angular and radial parts. Under such circumstances it is difficult to decide whether the errors in the latter calculation stem from the use of the Coulomb approximation or from errors in the intermediate-coupling coefficients (or both). Strong cancellations occur between the contributions of the 5*s* and 4*d* configurations in the calculations of some of the transition probabilities involved in these lifetimes; as a result, changes in either set of parameters could affect significantly the calculated lifetimes.

Our two estimates point out clearly that refined theoretical calculations in the Rb II are needed in order to match the accuracy of our experimental results. They also indicate that, despite their frequent use, estimates of lifetimes based upon semiempirical coupling calculations can be seriously in error.

ACKNOWLEDGMENTS

Two of us (M.G. and J.O.S., Jr.) wish to express our appreciation to the others for their hospitality during the course of the experimental work, and to the Fachbereich Physik and Sonderforschungsbereich 161 der Deutschen Forschungsgemeinschaft for financial support.

†Supported in part by the Fachbereich Physik of the Freie Universität Berlin and the Sonderforschungsbereich 161 der Deutschen Forschungsgemeinshaft.

[1]H. J. Andrä, A. Gaupp, K. Tillmann, and W. Wittmann, Nucl. Instrum. Methods 110, 453 (1973).
[2]H. J. Andrä, A. Gaupp, and W. Wittmann, Phys. Rev. Lett. 31, 501 (1973).
[3]H. Harde and G. Guthörlein, Phys. Rev. A 10, 1488 (1974).
[4]I. Martinson and A. Gaupp, Phys. Lett. 15C, 113 (1974).
[5]H. J. Andrä *et al.* (unpublished).
[6]*Handbook of Chemistry and Physics*, edited by R. C. Weast (Chemical Rubber, Cleveland, 1968–69), 49th ed.
[7]W. B. Bridges and A. N. Chester, Appl. Opt. 4, 573 (1965); A. R. Striganov and N. S. Sventitskii, *Tables of Spectral Lines of Neutral and Ionized Atoms* (I.F.I./Plenum, New York, 1968).
[8]J. Reader and G. Epstein, J. Opt. Soc. Am. 63, 1153 (1973).
[9]H. Kopfermann, A. Steudel, and J. O. Trier, Z. Phys. 144, 9 (1956).
[10]C. K. Kumar, G. E. Assousa, L. Brown, and W. K. Ford, Jr., Phys. Rev. A 7, 112 (1973).
[11]M. Aymar, S. Feneuille, and M. Klapisch, Nucl. Instrum. Methods 90, 137 (1970); M. Aymar, Physica 57, 178 (1972).
[12]M. Aymar and M. G. Schweighofer, Physica 67, 585 (1973).
[13]D. R. Bates and A. Damgaard, Philos. Trans. R. Soc. Lond. A 240, 101 (1949).
[14]G. K. Oertel and L. P. Shomo, Astrophys. J. Suppl. 147, 175 (1968).
[15]H. Statz, C. L. Tang, and G. F. Koster, J. Appl. Phys. 34, 2625 (1963); P. W. Murphy, C. L. Tang, and G. F. Koster, J. Opt. Soc. Am. 58, 1200 (1968).