Hyperfine Structure and Lifetimes of the $4^2P_{3/2}$ and $5^2P_{3/2}$ States of K^{39}†

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The hyperfine constants of the $4^2P_{3/2}$ and $5^2P_{3/2}$ states of K^{39} have been measured using the optical level-crossing method. The results are: $a(4^2P_{3/2}) = 6.0 \pm 0.1$ Mc/sec; $b(4^2P_{3/2}) = 2.9 \pm 0.2$ Mc/sec; $a(5^2P_{3/2}) = 1.95 \pm 0.05$ Mc/sec; $b(5^2P_{3/2}) = 0.92 \pm 0.1$ Mc/sec. These hfs constants are accurately proportional to the fine structure intervals to within 1%. These data, and the results from previous measurements on the $^2P_{1/2}$ states, yield values for the ratio $a(P_{1/2})/a(P_{3/2})$ in good agreement with measurements on other alkali atoms. The calculated quadrupole moment is $Q = (0.053 \pm 0.008) \times 10^{-24}$ cm^2, including Sternheimer corrections. The lifetimes of these states were determined to be: $\tau(4^2P_{3/2}) = 26.0 \pm 0.5$ nsec and $\tau(5^2P_{3/2}) = 140.8 \pm 1.0$ nsec.

I. INTRODUCTION

The hyperfine structure of the $4^2P_{3/2}$ and the $5^2P_{3/2}$ states of K^{39} are among the most difficult of the excited states of the alkali atoms to measure, primarily because the product of the hfs interaction energy and the natural lifetime is so small in each case. This product is important, since it measures the ratio of the hfs level separations to the natural width of the resonance line. If these quantities are comparable, the hfs levels are poorly resolved, and measurement of their separations is correspondingly more difficult. A handy rule that works quite well when the electric quadrupole interaction is small, and also for the alkali P states, is the following: The product of the magnetic dipole hfs coupling constant a(Mc/sec) and the lifetime τ(nsec) must be greater than 100 if a reasonable measurement is to be expected. Values of several hundred give good results, and values of a thousand or more can lead to precision measurements. For the $4^2P_{3/2}$ and $5^2P_{3/2}$ states of K^{39}, the values are roughly 153 and 277, respectively, and these are the smallest of any of the P states of the stable alkali isotopes, except possibly Li^7. This criterion is only a rough estimate; a more realistic one would involve the energy separations of the hfs levels. However, it is often correct, even if $b \sim a$, and it is easy to remember.

Only one direct measurement of the hfs interaction constants in the $4^2P_{3/2}$ state has been made,[1] using the atomic-beam magnetic-resonance method, and one measurement in the $5^2P_{3/2}$ state has been made,[2] using a double-resonance technique.

Indirect determinations are also possible. The procedure is to measure the hfs constant(s) in another state (which may have a more favorable resolution), and then relate that result to the state of interest by a theoretical formula. For instance, for K^{39}, a measurement of $a(5P_{1/2})$ can yield values for $a(5P_{3/2})$, $a(4P_{1/2})$, etc. However, the formulas may or may not be accurate, and in fact, what we will do is use the *direct* measurements to assess the accuracy of the formulas.

In this paper, we examine an inconsistency in the previous measurements on the quadrupole interaction constants in the $4^2P_{3/2}$ and $5^2P_{3/2}$ states of K^{39}, and present the results of a new and independent measurement of the hfs in these states. The new values remove the inconsistency, and allow a determination of the lifetimes of these states to a few percent, an improvement of an order of magnitude in the case of the $5^2P_{3/2}$ state.

II. DISCUSSION OF PREVIOUS MEASUREMENTS

The results of previous measurements in the first and second excited atomic states of K^{39} are listed in Table I. In the table, a is the magnetic dipole coupling constant and b is the electric quadrupole coupling constant. The Hamiltonian defining a and b is given by Buck and Rabi (Ref. 1).

From the Goudsmit-Fermi-Segre formula,[3] we can predict certain relationships between the entries of Table I. One of these is the ratio of the a factors within a fine structure doublet:

$$a(P_{1/2})/a(P_{3/2}) = 5.08,$$

where the value includes relativistic corrections but ignores the variation with principal quantum number n. This number is purely theoretical. From the results listed in Table I, we find

$$a(4P_{1/2})/a(4P_{3/2}) = 28.85/5.70 = 5.06 \pm 0.3,$$

$$a(5P_{1/2})/a(5P_{3/2}) = 8.99/1.97 = 4.56 \pm 0.3.$$

Table I. Previous measurements of hyperfine structure in excited states of K^{39}.

State	a (Mc/sec)	b (Mc/sec)
$4^2P_{1/2}$ [a]	28.85 ± 0.3	⋯
$4^2P_{3/2}$ [a]	5.70 ± 0.3	2.8 ± 0.8
$5^2P_{1/2}$ [b]	8.99 ± 0.15	⋯
$5^2P_{3/2}$ [c]	1.97 ± 0.1	1.7 ± 0.3

[a] P. Buck and I. I. Rabi, Phys. Rev. 107, 1291 (1957).
[b] W. N. Fox and G. W. Series, Proc. Phys. Soc. (London) 77, 1141 (1961).
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These values are slightly lower than the predicted values, a circumstance that has been observed in other alkali atoms[4] and associated with configuration interaction[5] and with core polarization resulting from valence-core exchange interaction.[6] It will be shown in Sec. IV that the new data yield ratios that are more consistent with the other measurements.

The theory also predicts that the hfs constants scale as the fine structure intervals, which for K^{39} are in the experimentally measured ratio

$$\delta\nu(5P)/\delta\nu(4P) = 18.76\ \text{cm}^{-1}/57.72\ \text{cm}^{-1} = 0.325.$$

Table I yields the following values:

$$a(5P_{1/2})/a(4P_{1/2}) = 8.99/28.85 = 0.312 \pm 0.06,$$

$$a(5P_{3/2})/a(4P_{3/2}) = 1.97/5.70 = 0.346 \pm 0.03,$$

$$b(5P_{3/2})/b(4P_{3/2}) = 1.7/2.8 = 0.635 \pm 0.22.$$

A discrepancy of nearly a factor of 2 is thus apparent in the ratio of the quadrupole coupling constants $b(5P_{3/2})$ and $b(4P_{3/2})$.

III. EXPERIMENT

The method of level-crossing spectroscopy was used in the present experiments. A functional diagram of the apparatus is shown in Fig. 1. Observation was made of the intensity of plane-polarized resonance radiation scattered at 90° by a collimated beam of free potassium atoms, versus the strength of an externally applied uniform magnetic field. The source of the resonance radiation was a microwave-excited electrodeless discharge lamp, made by distilling a small amount of potassium into a Pyrex tube $\frac{3}{8}$-in. diam and $1\frac{1}{2}$-in. long. The lamp was located in a coaxial cavity, and was viewed end-on. The light was incident on the atoms in a direction parallel to the magnetic field and was plane-polarized (perpendicular to the field). The scattered light was passed through an analyzer oriented perpendicular to the field, down a 1-m light pipe, and was detected by either a 7102 or 56 UVP photomultiplier in a thermoelectric cooler. The transverse components of the earth's field were canceled to less than 0.05 G with Helmholtz coils, and the longitudinal component of the earth's field, which does not affect the shape of the signals, was subtracted off in the analysis.

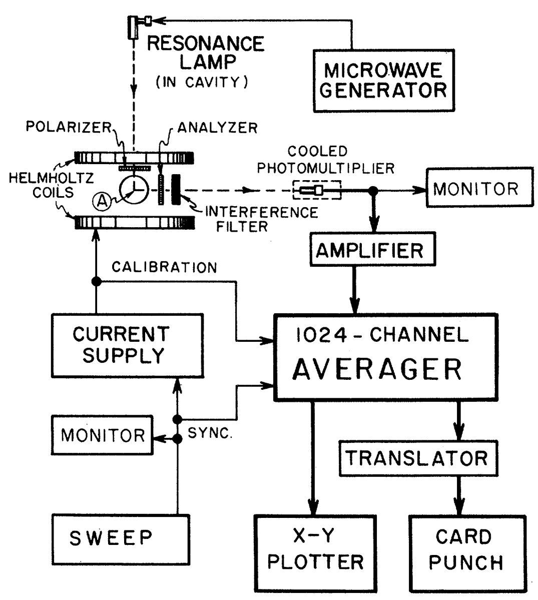

FIG. 1. Functional diagram of the apparatus. The light is scattered by a beam of free potassium atoms perpendicular to the drawing at point A. Not shown are the coils for cancelling the earth's magnetic field, lenses, vacuum system, etc.

The signal from the photomultiplier was fed into a 1024-channel signal averager. The magnetic field was swept in a triangular cycle with a 16-sec period, and the sweep of the averager was synchronized with the sweep of the field. The entire apparatus was very stable, and integration times of several hours, or several minutes per channel, were easily obtained. Stored data were transferred from the averager to punch cards by means of a BCD-to-decimal interface between the averager and the card punch. Data taking was sometimes interrupted for a readout, to insure against possible deterioration later.

The experimental data (intensity versus magnetic field) were compared with theoretical curves calculated from the Breit formula. The theoretical curves were generated by a program that diagonalizes the Hamiltonian to obtain the energy eigenvalues and eigenvectors for various values of the magnetic field, and then uses them in the Breit formula[7] to compute the scattered intensity. Figure 2 shows the energy levels versus magnetic field for the $P_{3/2}$ states of K^{39}. Both states behave similarly, so only the scales are different for the two states. The crossings with $\Delta M = 2$, which are observable in this experiment, are marked with a dot.

Implicit in the Breit formula is the assumption that the spectral intensity of the source is uniform over the absorption profile of the scattering atoms. This is certainly valid for K^{39}, where the ground-state hfs is small (461 Mc/sec) compared to the Doppler width of the lamp. Input parameters in the calculation are the hfs constants and the lifetime of the state. Comparison between the theoretical and experimental curves is made with a program that computes the mean square deviations between the two curves. The values of the parameters giving the best fit (least-mean-square deviation) are considered to be measured by the experiment. In the usual case, the hfs constants are well-known, so we obtain only the lifetime. In the case of the $5P_{3/2}$ state of K^{39} however, we were unable to get an acceptable fit to the experimental data for any value of the lifetime. Our procedure is described in the next section.

IV. DISCUSSION OF THE RESULTS

Since we were unable to fit the experimental data for the $4^2P_{3/2}$ state with the expected precision, we varied the hfs constants slightly. The best fit

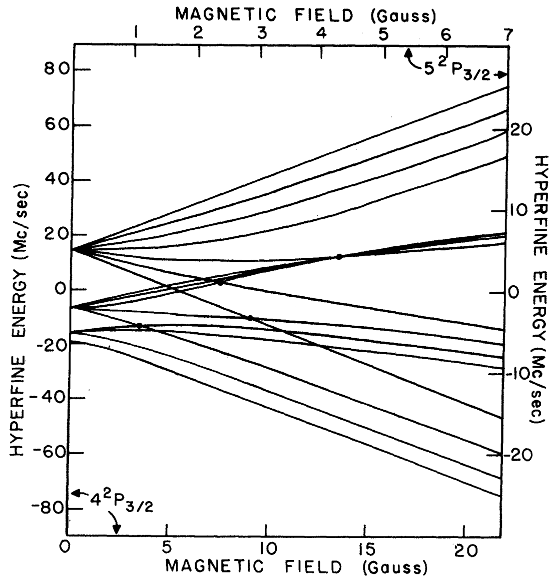

FIG. 2. Energy level diagram for the $^2P_{3/2}$ states of K^{39} considered in this paper. The observable crossings are marked. All the marked crossings are $\Delta M = 2$. The $F = 1$, $M = 0$ and the $F = 1$, $M = 1$ levels are inverted by a $\Delta M = 1$ crossing below 1 gauss.

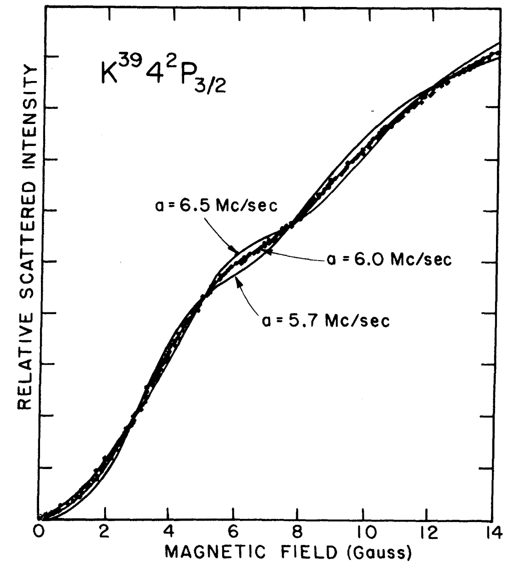

FIG. 3. Experimental and theoretical data for the $4^2P_{3/2}$ state of K^{39}. The theoretical curves are for the best lifetime $\tau = 26$ nsec, the best value of $b(4P_{3/2}) = 2.9$ Mc/sec, and three different values of $a(4P_{3/2})$.

was obtained using $a(4P_{3/2}) = 6.0 \pm 0.1$ Mc/sec and $b(4P_{3/2}) = 2.9 \pm 0.2$ Mc/sec. Figure 3 shows the experimental data, and theoretical curves for three sets of hfs constants. The best lifetime is $\tau(4P_{3/2}) = 26.0 \pm 0.5$ nsec, which agrees well with many other measurements.[8]

In attempting to fit the experimental data for the $5^2P_{3/2}$ state, we assumed $a = 1.97$ Mc/sec, $b = 1.7$ Mc/sec (cf. Table I), and varied the lifetime over a wide range (80-500 nsec). The mean square deviations showed no minimum, and the fits were very poor. Small changes in the hfs constants did not improve the fit appreciably, and we were led to consider the possibility that the hfs constants were seriously in error.

By assuming the hfs constants to be proportional to the fine structure, we could make the following predictions:

$$a(5P_{3/2}) = 0.325\ (6.0) = 1.95\ \text{Mc/sec},$$

$$b(5P_{3/2}) = 0.325\ (2.9) = 0.95\ \text{Mc/sec}.$$

The value 1.95 agrees well with the previously measured value of 1.97 (cf. Table I), but the b value, 0.95, is nearly a factor of 2 different. Thus the discrepancy in the ratio $b(5P_{3/2})/b(4P_{3/2})$ would seem to be almost entirely due to an incorrect value for $b(5P_{3/2})$. Following this prediction, we recalculated the theoretical curves, using several values for $b(5P_{3/2})$. The results are shown in Fig. 4, where a few of the curves for the best-fit lifetime have been plotted. The best-fit value of $b(5P_{3/2})$ is 0.92 ± 0.05 Mc/sec, which is not far from the predicted value of 0.95. Variation of the dipole hfs constant gave a best value $a(5P_{3/2}) = 1.95 \pm 0.05$ Mc/sec, in agreement with the predicted value and the previous experiment. It is readily apparent that the new values yield a much better fit over the entire curve. The best-fit lifetime is $\tau(5P_{3/2}) = 140.8 \pm 1.0$ nsec, which may be compared with the theoretical value[9] of 120 nsec and the only other measurement,[10] 379 nsec.

For convenience, we collect all the new results together in Table II. The errors represent two standard deviations. Using this data, and the data for the $P_{1/2}$ states from Table I, we get

$$a(4P_{1/2})/a(4P_{3/2}) = 28.85/6.0 = 4.81 \pm 0.15,$$

$$a(5P_{1/2})/a(5P_{3/2}) = 8.99/1.95 = 4.61 \pm 0.1,$$

$$a(5P_{3/2})/a(4P_{3/2}) = 1.95/6.0 = 0.325 \pm 0.015,$$

$$b(5P_{3/2})/b(4P_{3/2}) = 0.92/2.9 = 0.326 \pm 0.03.$$

It is fairly clear, then, that the hfs constants are accurately proportional to the fine structure, but that the relationship between $P_{1/2}$ and $P_{3/2}$ states is less well understood. It should be noted that the values listed in Table II are highly self-consistent, both in relation to the theory and with respect to other experiments.

The quadrupole moment is calculated from Ref.

Table II. Summary of the measurements described in this paper.

State	a (Mc/sec)	b (Mc/sec)	τ (nsec)
$4^2P_{3/2}$	6.0 ± 0.1	2.9 ± 0.2	26.0 ± 0.5
$5^2P_{3/2}$	1.95 ± 0.05	0.92 ± 0.1	140.8 ± 1.0

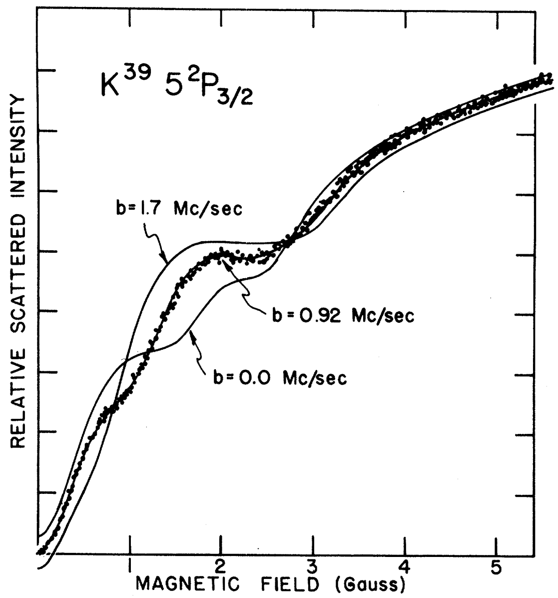

FIG. 4. Experimental and theoretical data for the $5^2P_{3/2}$ state of K^{39}. The theoretical curves are for the best lifetime $\tau = 140.8$ nsec, the best value of $a(5P_{3/2}) = 1.95$ Mc/sec, and various values of $b(5P_{3/2})$.

3 [Eq. (30.13), p. 156]. Using the new values for a and b in the $5^2P_{3/2}$ state, we get $Q = (0.057 \pm 0.004) \times 10^{-24}$ cm^2, which is in much better agreement with the value derived[1] from the $4^2P_{3/2}$ state, $Q = (0.07 \pm 0.02) \times 10^{-24}$ cm^2, than the value derived[2] from the $5^2P_{3/2}$ state using old a and b values, $Q = (0.11 \pm 0.02) \times 10^{-24}$ cm^2. The Sternheimer correction for the $5P$ state was calculated[11] for two wave functions, and there is a 12% difference between them. If we assume the correction is the average of these two, the corrected value for the quadrupole moment is $Q_{corr} = (0.053 \pm 0.008) \times 10^{-24}$ cm^2.

The experimental technique used in these measurements has the following advantages: (1) The level-crossing method involves none of the perturbations (Bloch-Siegert shifts, power broadening, multiquantum transitions, etc.) inherent in the double-resonance method. (2) dc detection (which measures scattered intensity) introduces no line shape distortions, as does the conventional phase-sensitive technique (which measures the derivative of the intensity). Distortions may occur in the latter case, if harmonics are present in the modulation. (3) The sweep of the magnetic field was cyclical, and the sweep period was small (16 sec) compared to the total integration time (10 000 sec). In this way, slow drifts (changes in lamp intensity, atomic density, etc.) automatically cancel. The sweep was a triangular wave, although this shape was not necessary, since the analysis automatically eliminates nonlinearities when the time is eliminated as a parameter. (4) The use of an atomic beam is clearly superior to a cell. The large integration times allowed us to operate at extremely low atomic density, well below levels required for trapping. (5) Finally, the analysis involves fitting a calculated line shape to a large number of experimental points (about 400). We are therefore confident that the fitting process is unique—that is, no other set of parameters would yield as good a fit.

We have made the same measurements and analysis on the $3^2P_{3/2}$ and $4^2P_{3/2}$ states of Na^{23}. The results are all in general agreement with previous work.

We would like to thank J. Clendenin for excellent programming assistance.

Note added in proof, Two other recent measurements of the quadrupole moment of the K^{39} nucleus have come to our attention. G. Sprott and R. Novick (private communication) have obtained an uncorrected value of $Q = (0.058 \pm 0.008) \times 10^{-24}$ cm^2 in a study of auto-ionization. W. E. Baylis [Ph. D. Thesis, Max Planck Institut für Physik und Astrophysik, Munich, 1967(unpublished)] obtained $Q = (0.056 \pm 0.022) \times 10^{-24}$ cm^2 using level crossing in the $4^2P_{3/2}$ state.

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