

Hyperfine Structure of K^{39} in the $4P$ State*†

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The atomic-beam magnetic resonance method has been used to investigate the hyperfine structure of the $4P$ state of K^{39} . The apparatus and method of observation were similar to those used in previous experiments on sodium, rubidium, and cesium. For K^{39} , the magnetic-dipole interaction constant in the $4P_{3/2}$ state is $a_1 = 28.85 \pm 0.3$ Mc/sec, the electric-quadrupole interaction constant in the $4P_{3/2}$ state is $b = 2.8 \pm 0.8$ Mc/sec, and the quadrupole moment is $(0.07 \pm 0.02) \times 10^{-24}$ cm².

INTRODUCTION

THIS paper describes an application of the atomic-beam magnetic resonance method¹ to the study of the hyperfine structure of the $4P$ excited state of K^{39} . The purpose of the experiment was to measure the nuclear electric quadrupole moment of the atom.

A brief account of this experiment was published previously.² Before that, a preliminary report³ on an application of the double-resonance optical method⁴ to the study of the $5P$ state of K^{39} was published. The value of the quadrupole moment of K^{39} found by the double-resonance optical method disagreed with our value. We wish to take this opportunity to express our regrets to the authors of that report for neglecting to cite their work in our brief preliminary report. Since our report, they have published⁵ a new value for the quadrupole moment which does not disagree with ours.

APPARATUS

The apparatus used in this experiment was described in a previous paper⁶ on Na^{23} ; therefore only the modifications will be discussed here.

The potassium beam was obtained by heating potassium metal in the oven to a temperature of about 225°C.

The source of resonance radiation was an Osram spectral lamp, containing potassium and an inert gas, operated at 20 volts and 0.8 amp ac. The lamp was surrounded by a water jacket to decrease the amount of self-reversal.

The rf current in the hairpin was kept constant over the frequency range under investigation by monitoring the voltage in the transmission line outside the vacuum with a General Radio 1800-A vacuum-tube voltmeter. For a constant current in the hairpin, the dependence

of the rf voltage on the frequency was obtained by directly calibrating the voltmeter against an rf ammeter in the transmission line. The voltage is approximately linear with frequency up to 70 Mc/sec since the hairpin acts like a pure inductance at such low frequencies.

In the experiments on sodium, rubidium, and cesium, a 2- by 8-mil tungsten ribbon was used as the detector. This ribbon contained large amounts of impurities and our mass spectrometer showed that the principle impurity was potassium. Consequently a new detector was built to reduce the large background noise from the detector itself. It consisted of two 2-mil "undoped" tungsten wires which were parallel and touching and presented to the beam a width of 4 mils. This arrangement was used because the only "undoped" tungsten immediately available was 2-mil wire.

The highest transition frequency in the $4P_{3/2}$ state of K^{39} is about 20 Mc/sec, and in this region the ground state rf effect⁶ is very large, twenty to fifty times as large as the excited state rf effect. The ground state rf effect is the transition between the ground state levels $F=2, m_F=-1$, and $F=2, m_F=-2$, and at a field of one gauss, this transition occurs at 0.7 Mc/sec. With our high rf power, the tail of this resonance is troublesome up to 30 Mc/sec. The effect is so large that fluctuations in beam intensity and in rf power are large enough to conceal the desired signal completely. Furthermore, the ground state rf effect is decreased when the light is turned on because the light defocuses some of the atoms which have been focused by the rf. This effect is also larger than the excited state rf effect and has fluctuations.

The ground state rf effect was eliminated completely by the following scheme. An unmodulated low-frequency oscillator, which consisted of a General Radio 1233-A power amplifier driven by a General Radio 805-C signal generator, was added to the apparatus. It was connected to the hairpin with a length of RG5-U cable which contained a 10 mh choke to prevent the high-frequency oscillator from feeding power into the low-frequency oscillator. (See Fig. 1.) This oscillator fed an rf current of 50 ma at a frequency of 1.5 Mc/sec into the hairpin. This served to scramble the populations in the $F=2, m_F=-1$ and $F=2, m_F=-2$ ground state levels (as well as the other ground state levels) and by

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† Submitted by Peter Buck in partial fulfillment of the requirements for the degree of Doctor of Philosophy in the Faculty of Pure Science, Columbia University.

¹ I. I. Rabi, *Phys. Rev.* **87**, 379 (1952).

² Buck, Rabi, and Senitzky, *Phys. Rev.* **104**, 553 (1956).

³ G. J. Ritter and G. W. Series, *Proc. Phys. Soc. (London)* **A68**, 450 (1955).

⁴ J. Brossel and F. Bitter, *Phys. Rev.* **89**, 308 (1952). J. Brossel and A. Kastler, *Compt. rend.* **229**, 1213 (1949).

⁵ G. W. Series, *Phys. Rev.* **105**, 1128 (1957).

⁶ Perl, Rabi, and Senitzky, *Phys. Rev.* **98**, 611 (1955).

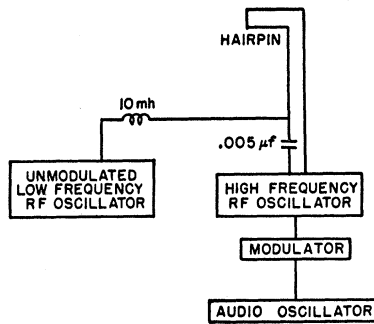


FIG. 1. Schematic drawing of the low-frequency oscillator and its associated equipment.

slight adjustments of current and frequency of the low-frequency oscillator, fifty percent of the atoms in the $F=2$, $m_F=-2$ ground state were transferred to the $F=2$, $m_F=-1$ state and vice versa. When the high-power rf was turned on, it also transferred atoms between these states but could not produce any additional refocused signal because the effect was already at a maximum. Consequently, the ground state rf effect was reduced to zero.

THEORY OF THE HYPERFINE STRUCTURE

The magnetic-dipole interaction constant a and the electric-quadrupole interaction constant b are defined by the Hamiltonian for the hyperfine structure energy at zero external field:

$$H(\text{hfs}) = ha \frac{C}{2} + hb \left[\frac{\frac{3}{2}C(C+1) - \frac{1}{2}I(I+1)J(J+1)}{IJ(2I-1)(2J-1)} \right], \quad (1)$$

where

$$C = F(F+1) - J(J+1) - I(I+1).$$

The transition frequencies with $\Delta F = \pm 1$ at near zero external field can be calculated from Eq. (1), and for

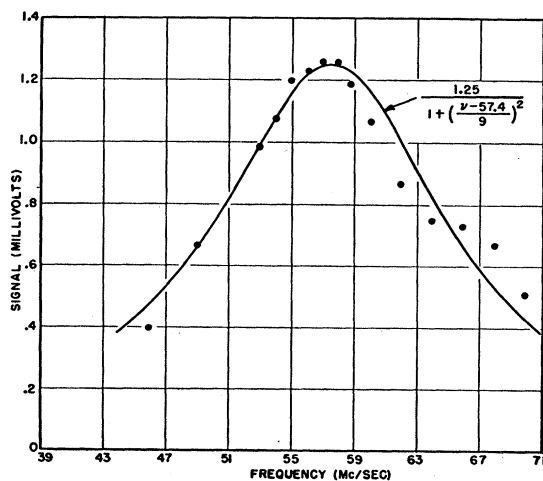


FIG. 2. The $F=2$ to $F=1$ transition in the $4P_{3/2}$ state of K^{39} .

TABLE I. Center frequency of the $F=2$ to $F=1$ transition in the $4P_{3/2}$ state of K^{39} .

Run	Observed center frequency in Mc/sec	Average in Mc/sec
1	57.5	57.7 ± 0.6
2	58.3	
3	57.4	

K^{39} are found to be

$$3a+b \quad \text{for } F=3 \text{ to } F=2,$$

$$2a-b \quad \text{for } F=2 \text{ to } F=1,$$

$$a-b \quad \text{for } F=1 \text{ to } F=0.$$

The interaction constants a and b may be calculated theoretically as⁷

$$a = \frac{2\mu_I\mu_0}{hI} \left(\frac{L(L+1)}{J(J+1)} \right) \mathcal{F} \left\langle \frac{1}{r^3} \right\rangle_{AV}, \quad (2)$$

$$b = - \left(\frac{2L}{2L+3} \right) R \left\langle \frac{1}{r^3} \right\rangle_{AV} Q, \quad (3)$$

where μ_I is the nuclear magnetic moment, μ_0 the Bohr magneton, and \mathcal{F} and R are relativistic corrections. Q is the electric quadrupole moment. From Eqs. (2) and (3),

$$Q = \frac{\mu_I\mu_0}{e^2} \left(\frac{(L+1)(2L+3)}{IJ(J+1)} \right) \frac{\mathcal{F}}{R} \frac{b}{a}. \quad (4)$$

EXPERIMENTAL RESULTS

A resonance was found at a frequency of 57.7 ± 0.6 Mc/sec which we assign to the transition $F=2$ to $F=1$ in the $4P_{3/2}$ state. The value of $a_{3/2}$ is then 28.85 ± 0.3 Mc/sec, which agrees well with the value 29.5 Mc/sec calculated from Eq. (2). Three runs were taken on this resonance and the results are shown in Table I. The rf spectrum of one of these runs is shown in Fig. 2, with a curve of the form $S = C \{1 + [(\nu - \nu_0)/\gamma]^2\}^{-1}$ superimposed. Here, S is the signal in millivolts, C a constant proportional to the gain of the detection system, γ the half-width at half-intensity, ν the applied frequency, and ν_0 the resonant frequency.

According to Eq. (2), the ratio $a_{3/2}/a_{1/2}$ is $5\mathcal{F}_{3/2}/\mathcal{F}_{1/2}$, where $\mathcal{F}_{3/2}$ and $\mathcal{F}_{1/2}$ are the relativistic corrections for the

TABLE II. Results of three runs on the $m_J = \frac{1}{2}$, $m_I = \frac{3}{2}$ to $m_J = -\frac{1}{2}$, $m_I = \frac{3}{2}$ transition in the $4P_{3/2}$ state of K^{39} .

Magnetic field in gauss	Observed center frequency in Mc/sec	$a_{1/2}$ in Mc/sec
81.8	161.0	5.6
79.6	158.0	6.1
79.7	157.0	5.4

⁷ H. B. G. Casimir, *On the Interaction between Atomic Nuclei and Electrons* (Teylers Tweede Genootschap, Haarlem, 1936).

$P_{3/2}$ and $P_{1/2}$ states, respectively. Casimir gives⁷ the values for these corrections for elements of various Z , and for potassium, with $Z=19$, the ratio $a_3/a_{3/2}$ is 5.14. Experimentally, however, we have found^{6,8} that the ratio is a few percent less than 5, with $a_3/a_{3/2}=4.95\pm 0.03$ for sodium and $a_3/a_{3/2}=4.8\pm 0.05$ for rubidium. We therefore take the ratio $a_3/a_{3/2}$ to be 5 for potassium and find a_3 to be 5.77 ± 0.06 Mc/sec. An error of a few percent in the ratio $a_3/a_{3/2}$ will not affect the final results importantly since the experimental uncertainty of the $P_{3/2}$ transition is so large.

An independent determination was made of the value of a_3 by investigating the Paschen-Back spectrum of the $4P_{3/2}$ state at a field of approximately 80 gauss. Twelve transitions are allowed with $\Delta m_J = \pm 1$, and $\Delta m_I = 0$. The rf effect due to the transition $m_J = \frac{1}{2}$, $m_I = \frac{3}{2}$ to $m_J = -\frac{1}{2}$, $m_I = \frac{3}{2}$ is nine times as strong as that resulting from any other transition and the Paschen-Back spectrum has one large peak due to this transition. The frequency of this transition is $g_J\mu_0 H/h + \frac{3}{2}a$ plus smaller terms in a and b which are much smaller than the experimental uncertainty. In Table II are listed the

TABLE III. Data on the $F=3$ to $F=2$ transition in the $4P_{3/2}$ state of K^{39} .

Run	Rf current in amp	Observed center frequency in Mc/sec	Half-width in Mc/sec
1	0.98	20.2	8.25
2	0.83	20.5	8.0
3	0.83	19.8	8.0
4	0.42	20.0	6.25

results of three runs on this transition. The value of a_3 found from these runs is 5.70 ± 0.27 Mc/sec which agrees with the value 5.77 ± 0.06 Mc/sec derived from a_4 .

Four runs were taken in the range from 15 to 30 Mc/sec and the rf spectrum from one of these runs is shown in Fig. 3. This spectrum was assumed to be the sum of two overlapping resonances due to transitions in the $4P_{3/2}$ state. This assumption was used to evaluate the center frequency of the upper resonance.

From Fig. 3, it can be seen that the rf spectrum is due to the superposition of a resonance at about 20 Mc/sec and one at a lower frequency at about 9 to 10 Mc/sec, both resonances having a half-width at half-intensity of about 8 Mc/sec. We make the assumption that the higher frequency resonance, f_1 , is due to the transition $F=3$ to $F=2$; therefore, $f_1=3a+b$. Since a is 5.77 Mc/sec, b equals about 3 Mc/sec. Then the transition $F=2$ to $F=1$ must lie at $2a-b$, which equals about 9 Mc/sec and is about 11 Mc/sec below the upper transition. With this preliminary estimate of the two center frequencies, curves of different relative heights and of different widths were added together and the resulting curve compared with the rf spectrum. The curve super-

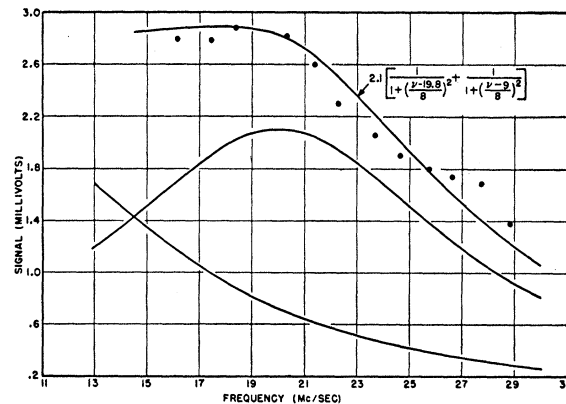


FIG. 3. The $4P_{1/2}$ rf spectrum with an rf current of 0.83^* amp in the hairpin. The higher frequency curve is assumed to be the $F=3$ to $F=2$ transition in the $4P_{3/2}$ state and the lower frequency curve is assumed to be the $F=2$ to $F=1$ transition. The top curve is the sum of the two lower curves.

imposed on the data in Fig. 3 is the best fit by this method and can be shifted 0.5 Mc/sec either way with reasonable agreement. The upper center frequency is quite independent of the lower center frequency and of the relative heights of the two curves used. Of course, for this very reason, little can be said about the lower center frequency. In Fig. 4, the rf spectrum from another run is shown. The rf current used in this run was half as great as that used in the run of Fig. 3 and the resonances are seen to be considerably narrower. The same curve-fitting procedure was used for these data and the half-width at half-intensity is 6.25 Mc/sec. This half-width is consistent with that of 8.0 Mc/sec for the first set of data and with the natural width of the line which is 5.9 Mc/sec. The natural width was obtained from the value for the half-life of the $4P_{3/2}$ state which is $(2.71\pm 0.09)\times 10^{-8}$ sec.⁹

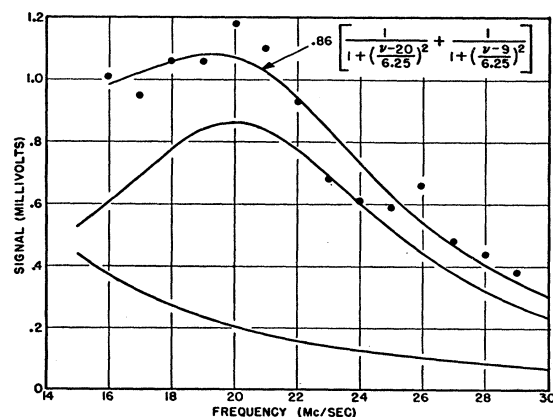


FIG. 4. The $4P_{1/2}$ rf spectrum with an rf current of 0.42 amp in the hairpin. The higher frequency curve is assumed to be the $F=3$ to $F=2$ transition in the $4P_{3/2}$ state and the lower frequency curve is assumed to be the $F=2$ to $F=1$ transition. The top curve is the sum of the two lower curves.

* B. Senitzky and I. I. Rabi, Phys. Rev. **103**, 315 (1956).

⁹ G. Stephenson, Proc. Phys. Soc. (London) **A64**, 458 (1951).

The results of four runs on the $F=3$ to $F=2$ transition are shown in Table III. The final value for the frequency of the $F=3$ to $F=2$ transition is 20.1 ± 0.4 Mc/sec, where the uncertainty quoted is the extreme deviation from the average. The value of b is then 2.8 ± 0.4 Mc/sec but, because of the difficulty of fitting

curves to the incompletely resolved resonances, we increase the uncertainty to ± 0.8 Mc/sec.

From Eq. (4), Q is found to be $(0.07 \pm 0.02) \times 10^{-24}$ cm².

We would like to thank Dr. Benjamin Senitzky for his help and advice during the course of the experiment.

Thermal-Neutron Fission Cross Sections for Isotopes of Plutonium, Americium, and Curium*

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The following thermal-neutron fission cross sections have been measured in the thermal column of the Materials Testing Reactor at Idaho Falls, Idaho: Pu²³⁸, 18.4 ± 0.9 barns; Am²⁴¹, 3.13 ± 0.15 barns; Am²⁴², 6390 ± 500 barns; Am²⁴³, < 0.072 barn; Cm²⁴³, 690 ± 50 barns; Cm²⁴⁵, 1880 ± 150 barns. In addition, a pile neutron capture cross section of 520 ± 40 barns has been measured for Pu²³⁸.

I. INTRODUCTION

THE increasing interest in the fission process and the value of knowing the destruction rate of certain nuclides during long neutron irradiations have made it desirable to determine accurately a number of slow-neutron fission cross sections for isotopes of the transuranic elements. It has become possible to perform some of these measurements because of the increased quantities of isotopes available, and because of the enrichments toward the heavier isotopes of certain elements gained in long neutron irradiations of Pu²³⁹ and Am²⁴¹. Fission cross-section measurements were made on Am²⁴², Am²⁴³, and Cm²⁴³ for which there were known previously either upper limits, approximate values or no data. In addition, the fission cross sections of Pu²³⁸, Am²⁴¹, Cm²⁴⁵, and the capture cross section of Pu²³⁸ were remeasured for purposes of comparison.

II. EXPERIMENTAL METHODS

Most of the samples were prepared from the transuranic elements separated from two long neutron irradiations in the Materials Testing Reactor, one of Pu²³⁹ (NR-3)¹ for a total integrated flux of 1.85×10^{22} neutrons/cm², and the other of Am²⁴¹ (99B)¹ for a flux of 4.08×10^{21} . After gross separation of the transuranium fraction from fission products, each element was highly purified by methods in common use at this laboratory which have been reported elsewhere.² In

each case, the final step of the chemical procedure yielded the pure element in acid solution from which nearly all foreign inorganic salts and organic impurities had been removed. The sample of Pu²³⁸ was separated as the alpha-decay product of 99B curium when its Cm²⁴² content was greater than 80%. The plates to be fission-counted were prepared by vacuum-vaporizing aliquots of the samples from hot tungsten filaments onto 1 in. \times 0.002 in. platinum disks. These plates were attached to a graphite shuttle which was then seated into an ionization type fission counter constructed of graphite and Lucite.³

The measurements of the fission counting rate were made in the thermal column of the Materials Testing Reactor in a neutron flux of approximately 5×10^{10} neutrons/cm² sec. The fission counter was the double chamber type with one chamber containing a Pu²³⁹ standard acting as a flux monitor. In the second chamber alternate counts were taken on the sample, blank background plates and Pu²³⁹ standards. Of the six Pu²³⁹ standards used, three were nearly isotopically pure Pu²³⁹, while the others contained a known amount of Pu²⁴⁰. The cross sections reported here are based on an 806-barn fission cross section for Pu²³⁹ for neutrons with a Maxwellian energy distribution in the thermal region.⁴ The counting rates at different discriminator voltages over the counter plateau were measured for each sample and an extrapolation to zero discrimination was made to normalize differences in plateau slope due to varying

* This work was performed under the auspices of the U. S. Atomic Energy Commission.

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¹ Arbitrary designations for samples obtained from particular irradiations.

² Thompson, Harvey, Choppin, and Seaborg, *J. Am. Chem. Soc.* **76**, 6229 (1954).

³ A. Ghiorso and W. C. Bentley, *The Transuranium Elements: Research Papers* (McGraw-Hill Book Company, Inc., New York, 1949), Paper No. 22.29, National Nuclear Energy Series, Plutonium Project Record, Vol. 14B, Div. IV.

⁴ *Neutron Cross Sections*, Atomic Energy Commission Report AECU-2040 (Technical Information Division, Department of Commerce, Washington, D. C., 1955), second edition.