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Natural Lifetimes and Hyperfine Structure for K^{39} in the $5p\,^2P_{3/2}$ and $6p\,^2P_{3/2}$ Levels of the K I Spectrum by Resonance Scattering of Light

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Abstract

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The hyperfine coupling constants and the natural radiative lifetimes of the $5p\,^2P_{3/2}$ and $6p\,^2P_{3/2}$ levels of K^{39} were determined in a level crossing experiment on an enriched K^{39} sample in a sealed-off resonance cell. The following results were obtained:

$a(5p\,^2P_{3/2}) = 1.973\ (12)$ MHz	$a(6p\,^2P_{3/2}) = 0.886\ (8)$ MHz
$b(5p\,^2P_{3/2}) = 0.870\ (18)$ MHz	$b(6p\,^2P_{3/2}) = 0.370\ (15)$ MHz
$\tau(5p\,^2P_{3/2}) = 133\ (3)$ ns	$\tau(6p\,^2P_{3/2}) = 310\ (15)$ ns

Considering the influence of core polarization on the magnetic hyperfine interaction and applying a Sternheimer correction, fairly consistent values of the electric quadrupole moment Q are obtained from the first three levels of the $np\,^2P_{3/2}$ sequence. We find $Q(K^{39}) = +0.059\ (6)$ barn.

1. Introduction

The optical double resonance and the level crossing techniques have been used in a large number of investigations of excited ^2P states of the alkali atoms. These high resolution methods have made possible a closer study of some effects not considered in the simplest theory for the alkali atoms. Core polarization effects have proved to have a considerable influence on the magnetic as well as the electric hyperfine structure [1–3]. Slight deviations from the Russell-Saunders value have been found for the Landé g_J factors [4–6]. For the study of different kinds of perturbations it is of great value to have experimental information from sequences of levels of the same type. So far investigations of more than two members in an alkali metal $np\,^2P_{3/2}$ series have been performed for Rb^{85}, Rb^{87}, Cs^{133} and Cs^{134} [1, 6–8]. The accuracy obtainable in an optical resonance experiment is determined by the ratio between the hyperfine structure splitting and the natural radiative width $\Gamma/2\pi = 1/2\pi\tau$ of the investigated level of lifetime τ. Contrary to the case for the heavy alkali metals Rb and Cs, resolved level crossing signals cannot be studied in level crossing experiments in $^2P_{3/2}$ levels of the lighter alkali metals. In such cases, the complicated signal structure obtained has to be interpreted in terms of coupling constants and natural radiative lifetime by a thorough comparison with theoretical curves calculated with the Breit formula [9], describing the process of resonance scattering.

For the $4p$ and $5p\,^2P_{3/2}$ levels of K^{39} two level crossing investigations have been performed, one by Schmieder, Lurio and Happer [10], and one by Ney [11]. The agreement between the magnetic dipole coupling constants a and the electric quadrupole coupling constants b obtained in these experiments is good, whereas there is a large discrepancy between the lifetime values τ for the $5p\,^2P_{3/2}$ level. The $4p\,^2P_{3/2}$ level has been studied by Buck and Rabi [12] using a modified optical double resonance technique, and in the $5p\,^2P_{3/2}$ level a double resonance investigation has been performed by Ritter and Series [13]. In the latter work a b factor 100% in error was obtained due to rf shifts not taken into account. This experiment has recently been reanalyzed yielding a b factor in accordance with the level crossing value [14]

In the present work the level crossing investigations of $^2P_{3/2}$ levels in K^{39} were extended to the third member of the $np\,^2P_{3/2}$ series. Because of the large discrepancy between the two level crossing lifetime values for $5p\,^2P_{3/2}$ this level was studied further. The nuclear quadrupole moment was calculated from the hyperfine structure of the different $^2P_{3/2}$ states. A fair agreement between the values was obtained after taking the influence of core polarization into account.

2. Resonance scattering from $^2P_{3/2}$ levels in K^{39}

The Hamiltonian used for the calculation of the energy levels of a $^2P_{3/2}$ level in the presence of hyperfine structure, as well as the Breit formula describing the process of resonance scattering have been presented and discussed in previous papers [15, 16]. In Fig. 1 the $^2P_{3/2}$ energy level diagram is reproduced for an atom with nuclear spin $I=3/2$ and $b/a=0.42$. As the energy is expressed in units of $h \cdot a$ and the magnetic field in units of $h \cdot a/g_J \cdot \mu_B$ the diagram represents an arbitrary member of the $^2P_{3/2}$ series provided that the ratio b/a is constant. For the geometric arrangement used in this experiment coherence effects between magnetic sublevels with $\Delta m=1$ are not obtained. The observed signal structure is produced by changes in the coherent scattering from $\Delta m=2$ magnetic sublevels and the change in the incoherently scattered light from each individual sublevel ($\Delta m=0$). Particularly strong effects are obtained close to the $\Delta m=2$ level crossings indicated in the Figure. Coherent scattering from two $\Delta m=2$ magnetic sublevels is only possible in the field region of mutual overlap within the radiation widths $\Gamma/2\pi=1/2\pi\tau$. Resolved level crossing signals can thus be obtained if the hyperfine splitting is large compared to $\Gamma/2\pi$. In the $^2P_{3/2}$ states of the heavy alkali metals, where resolved crossings are generally observed, the ratio $a/(\Gamma/2\pi)$ is of the order of 10. In the $4p$ and $5p\,^2P_{3/2}$ levels of K^{39} this ratio is very low, 1.00 and 1.65, respectively. In the

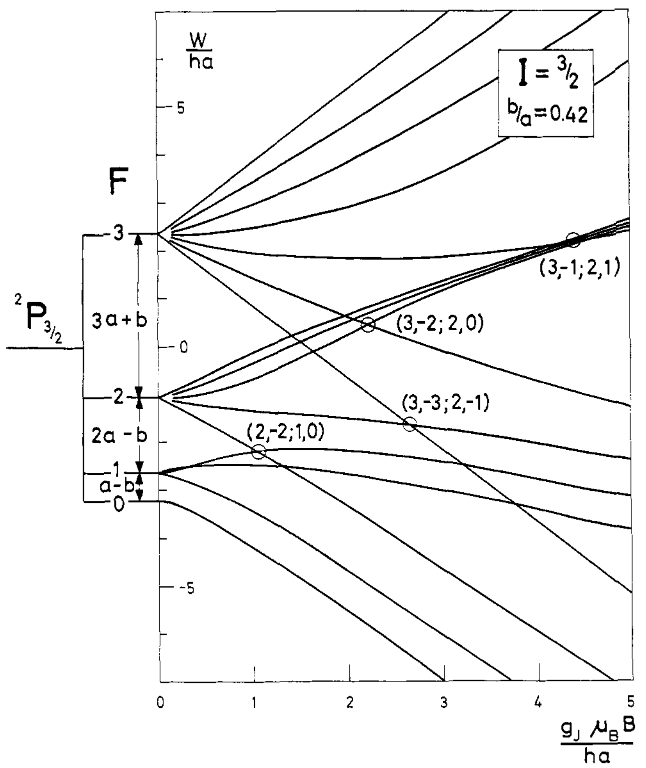

Fig. 1. Energy level diagram for a $^2P_{3/2}$ state in an atom with nuclear spin 3/2 and $b/a = 0.42$. The $\Delta m = 2$ level crossings are indicated using the symbol $(F, m_F; F', m_{F'})$.

$6p\ ^2P_{3/2}$ level the resolution does not improve considerably: the value 1.73 is obtained.

As the structure in the fluorescent radiation from excited $^2P_{3/2}$ levels in K^{39} is not very pronounced due to the low inherent resolution, it was favourable to enhance it by studying the derivative of the scattered intensity versus magnetic field as approximately obtained when the magnetic field is square wave modulated at a low frequency using the lock-in technique. For the analysis of this experiment a computer program, which calculates the scattered intensity according to the Breit formula for a given set of parameters a, b, g_J, and τ was used.[1] A direct comparison between the experimental curves and theoretical curves differentiated with the experimental magnetic field modulation could be done. In the calculation with the Breit formula, "white" excitation, i.e. a constant exciting light intensity over the wavelength region of absorption is assumed. For K^{39}, a spectral line connecting an excited $^2P_{3/2}$ level with the $^2S_{1/2}$ ground state consists of two hyperfine components due to the splitting of 462 MHz between the two ground state hyperfine levels, which is large compared to the excited state splitting. The Doppler width of the components is calculated to be 950, 1800, and 2100 MHz for the $4p$, $5p$, and $6p$ transitions, respectively. The increasing Doppler width results in a strong overlap of the spectral components, making the assumption of white excitation realistic for the $5p$ and $6p\ ^2P_{3/2}$ levels in K^{39}.

3. Experimental arrangement and measurements

The experimental set-up used in the potassium experiments has been described earlier [16]. For the excitation of the potassium atoms a sealed-off electrodeless rf lamp was used. The lamp cylinder, which was made of alkali resistent Osram "Überfang-Glas" Type 612 V 6/905c, was sealed off without any inert gas filling after a small amount of natural potassium (93% K^{39}, 7% K^{41}, and 0.01% K^{40}) had been distilled into it. In preliminary measurements natural potassium was used even in the Pyrex resonance cell. However, as the influence of K^{41} on the signal structure at low magnetic fields is not negligible, cells containing enriched K^{39} were produced.[2] About 20 mg metal, reduced from KCl with $Ba(N_3)_2$, was used in a cell. As the magnetic field region of interest is limited to the region 0 to 1.5 Gauss in the case of the $6p\ ^2P_{3/2}$ level, special care had to be taken in the compensation of the earth's magnetic field. The main Helmholtz coils used to generate a constant dc field, as well as the sweep and modulation coils, were calibrated by optical pumping in the ground state of K^{39}. At very low magnetic fields, calibration with K^{39} is advantageous compared to Cs^{133} and Rb^{85} which was used previously, since resolved resonances can be studied at much lower fields for K^{39} because of the smaller ground state hyperfine splitting. The current in the coil system compensating the vertical component of the earth's magnetic field was set by maximizing the current in the main coils necessary to achieve resonance for a fixed low radio frequency. The calibration of the square wave modulating field is illustrated in Fig. 2. The distance between the two peaks obtained by sweeping the main field through the resonance for an unmodulated rf field of low frequency directly gives the peak-to-peak modulation amplitude ΔB.

In the level crossing experiments the selection of the studied $^2P_{3/2}$ states was performed by means of narrow band interference filters in front of the detection EMI 9558 BQ photomultiplier. The suppression of other spectral lines was carefully investigated with a monochromator. Most of the measurements in the $5p$ and $6p\ ^2P_{3/2}$ levels were performed at a cell temperature of 80°C and 130°C, respectively, where the influence of coherence narrowing was found to be negligible. The differentiated signal as obtained from the lock-in amplifier was stored in a Laben Correlatron averaging multichannel analyzer, which controlled a repetitive magnetic field sweep.

A possibility to check the assumption of white excitation is given by observing the signals for different polarization combinations $[P_e, P_d]$ between the exciting and the detected light. Assuming white excitation, $[\sigma, \pi]$ and $[\pi, \sigma]$ curves should be identical and of half amplitude and opposite sign compared to $[\pi, \pi]$ curves [11]. In order to establish these different experimental testing situations, the exciting light, polarized with a Polacoat PL 40

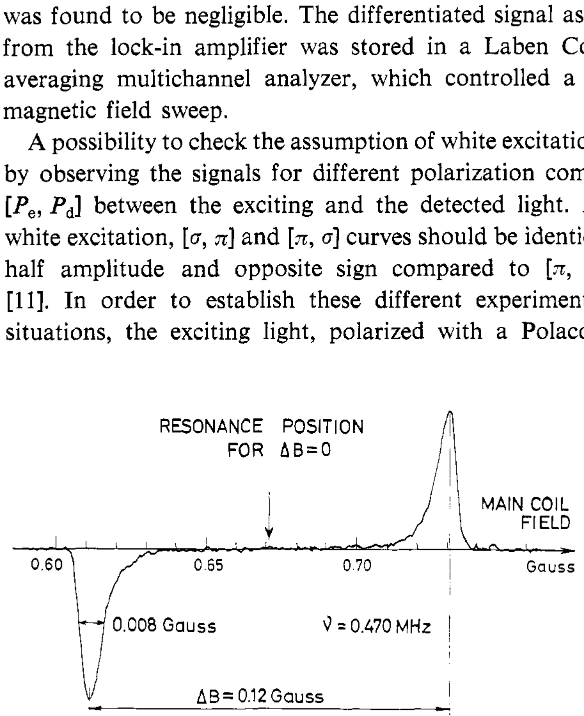

Fig. 2. Lock-in registration of optical pumping signals in the ground state of K^{39}, used to calibrate the square-wave modulation field. When ΔB is much larger than the signal half-width, the resonance condition is fulfilled for two different main coil fields, separated by ΔB. The undifferentiated signal, although somewhat disturbed by a not ideal square-wave form and the modulation coil inhomogeneity is obtained twice instead of the differentiated signal produced when a vanishing modulation amplitude is used.

[1] The author is grateful to Tekn. lic. G. Lidö and FK S. Rydberg for setting up and improving the Breit formula program due to W. Happer.

[2] The K^{39} sample delivered by Oak Ridge National Laboratory, Tenn., USA, had an isotopic purity of 99.97%.

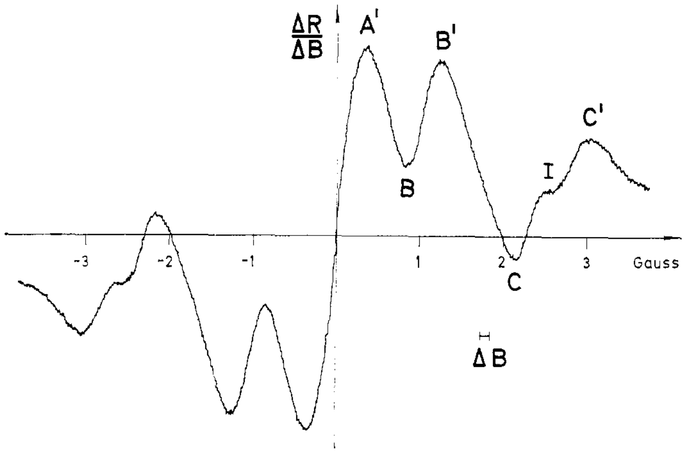

Fig. 3. Signal structure in the $5p\ ^2P_{3/2}$ level obtained by adding 120 magnetic field sweeps in the multichannel analyzer. The duration of each sweep was 60 s. and the lock-in time constant was set to 0.1 s. Peak-to-peak modulation amplitude: 0.12 Gauss.

Table I. *[σ, σ] scattering results for the $5p\ ^2P_{3/2}$ and the $6p\ ^2P_{3/2}$ levels*

Full modulation amplitude: 0.12 Gauss. Three standard deviation errors

	A′	B	B′	C	I	C′
$5p\ ^2P_{3/2}$ $\lambda = 4\,044$ Å						
Position, Gauss	0.352 ±0.004	0.846 ±0.007	1.246 ±0.004	2.147 ±0.008	2.569 ±0.014	3.040 ±0.015
Intensity, Arbitr. units	34.5 ±0.4	12.7 ±0.5	31.7 ±0.4	−3.7 ±0.4	8.5 ±0.3	17.4 ±0.3
$6p\ ^2P_{3/2}$ $\lambda = 3\,446$ Å						
Position, Gauss	0.159 ±0.006	0.386 ±0.006	0.576 ±0.006	0.960 ±0.005	— —	1.370 ±0.010
Intensity, Arbitr. units	36.9 ±2.1	12.2 ±1.5	31.1 ±0.6	−3.1 ±1.1	— —	16.7 ±0.9

filter, was irradiated onto the cell at right angles to the magnetic field, and the fluorescent light was detected through a PL 40 filter at right angles to the magnetic field as well as to the incident light direction. For a lamp temperature of 160°C and an rf input power of 100 W, curves obtained for different polarizer settings corresponded very well to a situation of white excitation. For the [σ, σ] registrations the light source was sometimes moved and σ-light was irradiated through a PL 40 polarizer in the direction of the magnetic field and the σ-component of the fluorescent light was detected. This arrangement was found to give a more favourable signal-to-noise ratio than the other arrangement.

The [σ, σ] scattering signal structure for the $5p$ and the $6p$ $^2P_{3/2}$ levels is reproduced in Figs. 3 and 4 as obtained with the lock-in technique when a square-wave magnetic field modulation of 0.12 Gauss peak-to-peak was used. The curves were obtained by adding the signal information from 120 and 90 magnetic field sweeps, respectively, using a sweep duration of 60 s and a lock-in time constant of 0.1 s. As nearly the same inherent resolution $a/(\Gamma/2\pi)$ is found for the two states, and as the ratio b/a is approximately constant, essentially a scale factor for the magnetic field connects the two structures. For identification, the peaks of the curves are labelled with A′, B, B′, C, and C′. The inflexion point I, generated between the second and the third distorted, strongly overlapping level crossing signals, is usable for evaluation only in the $5p\ ^2P_{3/2}$ level. In the $6p\ ^2P_{3/2}$ level it is not distinct because the modulation amplitude is comparatively larger

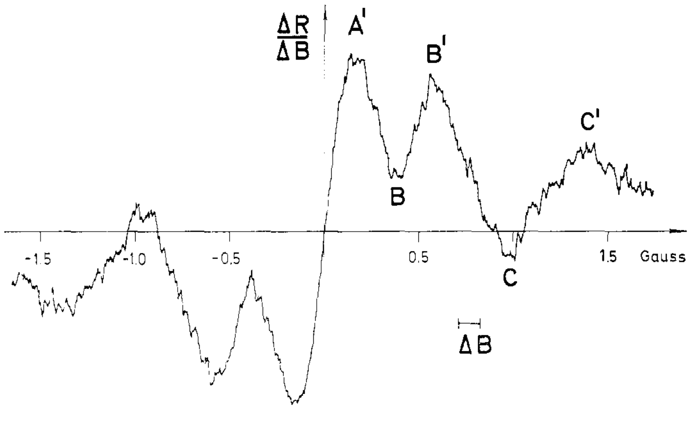

Fig. 4. Signal structure in the $6p\ ^2P_{3/2}$ level obtained by adding 90 magnetic field sweeps. Other data as in Fig. 3.

and a worse signal-to-noise ratio is obtained. In order to determine the magnetic field values of the characteristic points, a very short magnetic field region centered around the single points was swept for increasing and decreasing fields as described in Ref. [16]. In order to determine the relative intensities of the different peaks, over-all runs like those in Figs. 3 and 4 were made. The experimental data obtained from 6 and 8 measuring series for the $5p$ and $6p\ ^2P_{3/2}$ level, respectively, are reproduced in Table I. The measurements were done on several different occasions, and in each case an optical adjustment and a symmetrization of the signal curve as well as a polarization test on the lamp burning conditions were undertaken.

4. Determination of a, b, and τ

Under the assumption of white excitation the signal structure obtained in a resonance scattering experiment is determined by the quantities a/g_J, b/g_J, and $\tau \cdot g_J$. For the $^2P_{3/2}$ level g_J factor, slight deviations from the LS-value -1.3341 have been found for Cs and Rb. However, the deviation in no case exceeds 0.2%, and for the accuracy obtained in this experiment the LS-value is accurate enough for obtaining the values of a, b, and τ. With the Breit formula computer program, the signal structure obtained for the experimental modulation amplitude is calculated for different combinations of a, b, and τ. It is found that the point C to a large extent is insensitive to variations in b and τ, whereas its magnetic field position is proportional to a. Thus it is fairly easy to get a good starting value of a in the fit of a theoretical curve to the experimental curve. For the determination of b the magnetic field values of the peaks B′ and C′ are suited as they are comparatively little influenced by τ. The point A′ is dependent almost only on τ. The shape of the curve around the inflexion point I is also very sensitive to τ. The relative intensities of the peaks are closely related to the values of τ and b. The final fit is found by varying all parameters with special attention to the behaviour of the theoretical curve in points where the influences of the parameters to a large extent are "orthogonal". In the two $^2P_{3/2}$ levels the best fit was obtained with the following parameters:

$a(5p\ ^2P_{3/2}) = 1.973(12)$ MHz $\quad a(6p\ ^2P_{3/2}) = 0.886(8)$ MHz

$b(5p\ ^2P_{3/2}) = 0.870(18)$ MHz $\quad b(6p\ ^2P_{3/2}) = 0.370(15)$ MHz

$\tau(5p\ ^2P_{3/2}) = 133(3)$ ns $\quad \tau(6p\ ^2P_{3/2}) = 310(15)$ ns

Table II. *Comparison between results for the 4, 5, and 6p $^2P_{3/2}$ levels in K^{39}*

Level	a MHz	b MHz	τ ns	Ref.
$4p\ ^2P_{3/2}$	6.13 (5)	2.72 (12)	28 (2)	Ney [11]
$\frac{a}{\Gamma/2\pi}\approx 1.00$	6.0 (1)	2.9 (2)	26.0 (0.5)	Schmieder et al. [10]
	5.7 (3)	2.8 (8)		Buck et al. [12]
			26.9	Heavens [17]
$5p\ ^2P_{3/2}$	1.97 (2)	0.85 (3)	120 (4)	Ney [11]
	1.95 (5)	0.92 (10)	140.8 (1.0)	Schmieder et al. [10]
$\frac{a}{\Gamma/2\pi}\approx 1.65$	1.97 (10)	~ 1		Ritter et al. [13] Pegg [14]
			120	Heavens [17]
	1.973 (12)	0.870 (18)	133 (3)	This work
$6p\ ^2P_{3/2}$			300	Heavens [17]
$\frac{a}{\Gamma/2\pi}\approx 1.73$	0.886 (8)	0.370 (15)	310 (15)	This work

In the quoted errors the statistical error in the determination of the peaks (three standard deviations) as well as the estimated uncertainty in the fitting procedure was taken into account.

5. Comparison with previous results

The available data from the experiments mentioned in the introduction are given in Table II together with the results from this experiment. The a and b factors of the $5p\ ^2P_{3/2}$ level obtained in the present experiment are in good agreement with the previous results. For the lifetime a value between the two previous, mutually inconsistent values is obtained. For the first three $^2P_{3/2}$ levels the values calculated by Heavens [17] using the Coulomb approximation are in good agreement with the experimental results.

6. Core polarization and calculation of Q

In order to obtain a reliable value of the nuclear quadrupole moment Q from the measured coupling constants, it is necessary to take the influence of core polarization effects on the magnetic dipole and the electric quadrupole interaction into account. In a recent review article Fischer has discussed such influences on the hyperfine structure of alkali-like atoms [2]. Following the approach previously used by zu Putlitz [1] and Hartmann [18] for Rb and Na, respectively, we can calculate the contribution a_c to the dipole interaction, due to core polarization. Using

Table III. *Calculation of the influence of core polarization on the magnetic hyperfine structure of ^2P states in K^{39}*

$$\left(F_r(3/2)=\frac{1.002}{(1.0047)},\ F_r(1/2)=\frac{1.030}{(1.0224)}\right)$$

	$a_{3/2}$ [a] MHz	$a_{1/2}$ MHz	$a_{3/2\,\mathrm{corr}}$ MHz	$\frac{a_c}{a_{3/2}}$
$4p\ ^2P$	6.09 (4)	28.85 (0.3) [b]	5.69 (5.74)	0.066 (0.057)
$5p\ ^2P$	1.972 (10)	8.99 (15) [c]	1.785 (1.801)	0.095 (0.087)
$6p\ ^2P$	0.866 (8)	—	0.814 (0.822)	0.081 (0.072)

[a] Weighted mean values from Table II.
[b] Ref. [12]. [c] Ref. [21].

Table IV. *Calculation of $\langle a_0^3/r^3\rangle$.* ($F_r(3/2)=1.002$, $H_r=1.0066$)

	From $a_{3/2}$	From $a_{3/2\,\mathrm{corr}}$	From δW ($Z_i=16.7$)
$4p\ ^2P$	0.458	0.428	0.392
$5p\ ^2P$	0.148	0.134	0.127
$6p\ ^2P$	0.0666	0.0612	0.0571

the experimental dipole coupling constants $a_{3/2}$ and $a_{1/2}$ for the $^2P_{3/2}$ and $^2P_{1/2}$ levels, respectively, the corrected values $a_{3/2\,\mathrm{corr}}=a_{3/2}-a_c$ and $a_{1/2\,\mathrm{corr}}=a_{1/2}+a_c$, referring only to the valence p-electron are obtained. The result is for the heavy alkali metals very sensitive to the relativistic correction factors used. As pointed out by Lindgren and Rosén [19] the factors given by Kopfermann [20] can be quite uncertain. Lindgren and Rosén have calculated new values for the correction factors $F_r(3/2)$, $F_r(1/2)$, and R_r by comparing the values of $\langle a_0^3/r^3\rangle$ obtained in relativistic and nonrelativistic self-consistent-field calculations.

In Table III mean values of the $a_{3/2}$ factors obtained by weighting the different experimental results inversely proportional to the square of the errors are given together with the experimental $a_{1/2}$ factors for the 4 and $5p\ ^2P_{1/2}$ levels [12, 21]. The corrected values $a_{3/2\,\mathrm{corr}}$ and the ratios $a_c/a_{3/2}$ as calculated with the new relativistic correction factors and with the old ones (in parentheses) are also given in the Table. As relativistic effects are small for light elements as K^{39} the results are little affected by the choice of factors. Assuming the contribution from the core polarization (a_c) to be a constant fraction of $a_{3/2}$ [2], $a_{3/2\,\mathrm{corr}}$ for the $6p\ ^2P_{3/2}$ level could be obtained despite the lack of an experimental $a_{1/2}$ factor. For this purpose the mean value $a_c/a_{3/2}=0.081$ (0.072) from the $4p$ and $5p$ levels was used. In Table IV values of $\langle a_0^3/r^3\rangle$ are calculated from $a_{3/2}$, $a_{3/2\,\mathrm{corr}}$, and from the fine structure splitting δW, using the one-electron formulas given in Ref. [20]. The new correction factors have been used. In the fine structure calculation we have used the effective nuclear charge $Z_i=16.7$, obtained in the self-consistent-field calculations by Lindgren and Rosén [19]. This value is to be compared with $Z_i=Z-4=15$ frequently used [20], and $Z_i=17.3$ obtained by Sternheimer et al. [22]. The discrepancy between the two last columns in Table IV is not surprising with regard to the simplified approach with which the values were obtained, e.g., we have not considered two-particle effects in the fine structure [23].

For the calculation of the nuclear electric quadrupole moment Q, weighted mean values of the experimental b factors in each level and the mean values $\langle a_0^3/r^3\rangle_{\mathrm{mv}}$ of the corrected hyperfine structure values and the fine structure values from Table IV were used. The values of Q given in the fourth column of Table V decrease in the $np\ ^2P_{3/2}$ level sequence. Sternheimer has performed several calculations on the shielding or antishielding of the quadrupole moment by the core electrons. In a recent work [3] the correction

Table V. *Calculation of the quadrupole moment.* ($R_r=1.007$)

Level	b^a MHz	$\left\langle\frac{a_0^3}{r^3}\right\rangle_{\mathrm{mv}}$	Q barn	$\frac{1}{1-R}$ [b]	Q_{corr} barn
$4p\ ^2P_{3/2}$	2.77 (10)	0.410	0.0714	0.824	0.0588
$5p\ ^2P_{3/2}$	0.866 (15)	0.131	0.0699	0.853	0.0596
$6p\ ^2P_{3/2}$	0.370 (15)	0.0591	0.0661	0.863	0.0570

[a] Weighted mean values from Table II.
[b] Ref. [3].

factors $1/(1-R)$ given in the Table were obtained. The application of these correction factors leads to a reduction of the quadrupole moment with some 15% and an improved consistency between the values of Q as obtained from the different $^2P_{3/2}$ levels is found. The uncertainty in the quadrupole moment due to the determination of b is about 4, 2, and 4% in the 4, 5, and $6p\ ^2P_{3/2}$ levels, respectively. However, the final uncertainty in the quadrupole moment is considerably larger due to the uncertainties in the evaluation of $\langle a_0^3/r^3\rangle$ and in the calculation of the Sternheimer correction factors. Estimating the possible errors to be 8% and 5%, respectively, we obtain for the nuclear quadrupole moment of K^{39} $Q(K^{39}) = -0.059(6)$ barn.[1]

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[1] Applying the present analysis to the experimental results obtained for K^{40} in a previous work [15], the quadrupole moment of K^{40} is found to be $Q(K^{40}) = -0.075\ (11)$ barn.