PHYSICAL REVIEW A

GENERAL PHYSICS

THIRD SERIES, VOL. 3, NO. 3 MARCH 1971

Quadrupole Antishielding Factors, and the Nuclear Quadrupole Moments of Several Alkali Isotopes*

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(Received 2 October 1970)

The atomic antishielding factor R for the quadrupole hyperfine structure has been evaluated for the lowest three excited np states for each of the alkali atoms, i.e., for a total of fifteen atomic states. The values of R were determined by means of the perturbed wave functions $v_1'(nl\to l')$, as obtained by solving the inhomogeneous Schrödinger equation for each type of excitation of the core electrons by the nuclear quadrupole moment Q. In order to obtain reliable values of R, we have obtained accurate valence wave functions $v(np)$ for the np excited states from the requirement that the calculated energy eigenvalues $E_0(np)$ should reproduce the observed energy levels. The wave function $v(n_0p)$ for the lowest valence np state ($n=n_0$, e.g., $n_0=3$ for Na) was checked for each alkali atom by calculating the oscillator strength $f_{n_0s-n_0p}$ for the lowest $n_0s\to np$ transition. The calculated values of $f_{n_0s-n_0p}$ were found to be in good agreement with experiment. As an example of the values of $R(np)$ obtained, we have found for Rb: $R(5p)=-0.257$; $R(6p)=-0.207$; and $R(7p)=-0.190$. The resulting correction factors $\mathcal{C}=1/(1-R)$ are $\mathcal{C}(5p)=0.796$, $\mathcal{C}(6p)=0.829$, and $\mathcal{C}(7p)=0.840$. At the end of the paper we give the corrected values of Q for twelve alkali isotopes, for which experimental data are available, namely, for the following nuclei: Na^{23}, K^{39}, K^{40}, K^{41}, Rb^{85}, Rb^{87}, Cs^{131}, Cs^{132}, Cs^{133}, Cs^{134}, Cs^{135}, and Cs^{137}. We have also given a brief summary of the present experimental evidence supporting the existence of the quadrupole shielding and antishielding effects for atomic states.

I. INTRODUCTION

The purpose of the present paper is to give the results of a systematic calculation of the quadrupole antishielding factor[1] R for the first three excited np states of each of the alkali atoms. The resulting values of R are then used to obtain corrected values of the nuclear quadrupole moments Q for these elements, as derived from spectroscopic measurements of the quadrupole hyperfine structure (hfs) in the excited np states. In the course of this investigation, we have obtained accurate valence wave functions for the np electrons from the requirement that the energy eigenvalues should reproduce the observed energy levels.[2] At the end of Sec. II, these wave functions are used to obtain accurate values of the oscillator strengths f_{n_0s-np} for transitions from the ground state n_0s. It is shown that these oscillator strengths are in good general agreement with the corresponding measured values for the transition to the lowest np state, i.e., $f_{n_0s-n_0p}$. Here n_0 is the principal quantum number for the ground state of the valence electron, e.g., $n_0=3$ for Na. We note that in obtaining the f_{n_0s-np}, we have employed the ground-state wave functions $v_0(n_0s)$ for the alkali atoms, which were previously determined in a calculation of the alkali atom polarizabilities.[3]

The present calculations of R for the alkali atoms constitute a substantial improvement over those carried out previously by one of us (R. M. S.)[4] in 1956. Thus the exchange terms with the valence electron are included in the present calculations (as opposed to the earlier ones); the individual angular perturbations of the core $v_1(nl\to l+2)$ were calculated in each case (instead of relying on the less accurate Thomas-Fermi model), and, more importantly, we have determined and employed np valence wave functions $v(np)$ which reproduce the observed atomic energy levels[2] for these states. The resulting values of R confirm the existence of the antishielding obtained previously.[1,4] However, the estimated uncertainty of the present results is considerably reduced, as compared to those of the earlier calculations.

II. CALCULATIONS OF R

The formalism used to calculate R is essentially

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the same as that employed in our earlier papers on this subject,[1,4,5] in particular, Ref. 5, in which we have presented the results of a calculation of R for two states of the Cu atom ($3d^9 4s^2$ and $3d^{10}4p$), and for Pr, Tm, Li, and Be. In particular, it was shown in Ref. 5 that the two very different values of Q for the copper isotopes as derived from $3d^9 4s^2$ and $3d^{10}4p$ (a difference of approximately 40%) can be brought into very good agreement by applying the corresponding correction factor $\mathfrak{C} \equiv 1/(1-R)$, which are quite different for the two configurations, namely, $1/0.822 = 1.217$ for $3d^9 4s^2$ and $1/1.175 = 0.851$ for $3d^{10}4p$.

The radial wave functions $v_1'(nl \to l')$ which describe the excitation of the core electrons (nl) by the potential due to the nuclear quadrupole moment Q are the solutions of the following inhomogeneous equation[1]:

$$\left(-\frac{d^2}{dr^2}+\frac{l'(l'+1)}{r^2}+V_0-E_0\right)v_1'(nl\to l') = u_0'(nl)\left(r^{-3}-\langle r^{-3}\rangle_{nl}\delta_{ll'}\right) , \tag{1}$$

where u_0' is r times the radial part of the unperturbed (nl) core wave function. The effective values[6] of $V_0(r) - E_0$ (V_0 is the unperturbed potential; E_0 is the unperturbed energy eigenvalue) were obtained in the same manner as in Refs. 1 and 6:

$$V_0 - E_0 = \frac{1}{u_0'}\frac{d^2 u_0'}{dr^2} - \frac{l(l+1)}{r^2} . \tag{2}$$

The method of solution of Eq. (1) has been described in Ref. 5 (see the discussion on pp. 13–15). The core wave functions $u_0'(nl)$ are the Hartree-Fock wave functions for the corresponding alkali (positive) ions. Thus for Na$^+$ and K$^+$, we used the functions of Hartree and Hartree[7]; for Rb$^+$ and Cs$^+$, the wave functions of Freeman and Watson[8] were employed. For Li, the $1s$ function of James[9] was used.

As mentioned in the Introduction, the valence wave functions $v(np)$ were determined from the requirement that they should reproduce the observed energy eigenvalues $E_0(np)$, i.e., the atomic energy levels obtained from the tables of Moore.[2] In several previous papers on the alkali atom polarizabilities,[3] one of us (R. M. S.) has used potentials which reproduce for each case the eigenvalue for the lowest $n_0 s$ (ground) state, i.e., the observed ionization potentials.[2] These effective potentials $V_{n_0 s}$ were obtained for Li, by Seitz[10]; for Na, by Prokofjew[11]; for Rb, by Callaway and Morgan[12]; and for K and Cs, by Sternheimer.[13,14] The potentials $V_{n_0 s}(r)$ should form a very good first approximation to the effective potentials for the excited np states, to be denoted by $V_{np}(r)$. That $V_{n_0 s}$ will not exactly reproduce the observed $E_0(np)$ is presumably due to the fact that the effects of exchange and correlation are slightly different for different valence states. For the purpose of obtaining the radial dependence (shape) of $\delta V \equiv V_{np} - V_{n_0 s}$, we have assumed that this dependence is given by the Slater exchange potential.[15] When this potential is multiplied by the factor $\frac{2}{3}$ introduced by Gáspár and by Kohn and Sham,[16] it is given by

$$V_{\mathrm{exch}}(r) = -4\left[(3/8\pi)\rho\right]^{1/3}\ \mathrm{Ry} , \tag{3}$$

where ρ is the electron density in atomic units (number of electrons $/a_H^3$, where a_H is the Bohr radius). In terms of the wave functions $u_0'(nl)$, which are normalized to 1,

$$\int_0^\infty [u_0'(nl)]^2 dr = 1 , \tag{4}$$

$V_{\mathrm{exch}}(r)$ can be written as follows:

$$V_{\mathrm{exch}}(r) = -\left(\frac{12}{\pi^2}\right)^{1/3}\left(\sum_{nl}\frac{(2l+1)[u_0'(nl)]^2}{r^2}\right)^{1/3}\ \mathrm{Ry} , \tag{5}$$

where the numerical factor $(12/\pi^2)^{1/3} = 1.0673$.

The potentials[17] $V_{n_0 s}(r)$ have been tabulated in Ref. 17, which pertains to the calculation of the alkali atom dipole and quadrupole polarizabilities[3] α_d and α_q. With the experimental value[2] of $E_0(np)$, we now write

$$V_{\mathrm{eff}}(np) = V_{n_0 s} + a\,|V_{\mathrm{exch}}| , \tag{6}$$

where a is an adjustable parameter determined by the Schrödinger equation for $v(np)$, namely,

$$\left(-\frac{d^2}{dr^2}+\frac{2}{r^2}+V_{n_0 s}+a\,|V_{\mathrm{exch}}| - E_0(np)\right)v(np) = 0 . \tag{7}$$

Thus $|V_{\mathrm{exch}}|$ provides the shape of the small correction to the potential $V_{n_0 s}$. Equation (7) is an eigenvalue equation for a, i.e., the constant a must be so chosen that $v(np)$ will be well behaved at $r \to \infty$. The computer program, which integrates Eq. (7), tries a large number of a values, and by interpolating between two final functions (both of which diverge very slowly as $r \to \infty$, with different signs), one obtains a function which behaves properly, i.e., it is approximately proportional to $\exp[-|E_0(np)|^{1/2} r]$ at large r. It may be noted that the total running time is 24 sec on a CDC-6600, distributed into three calculations of 8 sec each. Between each two successive calculations, one narrows down the range of a values which the computer investigates by a factor of ~ 50. Thus initially a is allowed to range from -0.5 to $+0.5$ (interval equal to 1), and after the third (final) set of integrations, a is determined to 10^{-5}. The values of a are quite small, as would be expected, since $V_{n_0 s}$ is a good approximation to the actual effective potential V_{np}. Thus $|a|$ is less than 0.15 in all cases. Another way of demonstrating the smallness of the correction term $a\,|V_{\mathrm{exch}}|$ is to calculate the energy which corresponds to it, namely,

$$\delta E = a\int_0^\infty |V_{\mathrm{exch}}|\, v_{np}^2\, dr , \tag{8}$$

and to compare δE to the total binding energy of the np electron[2] $|E_0(np)|$. In this connection, the integral of Eq. (8), to be denoted by $-E_{\text{exch}}(np)$, is tabulated in Table I for the first three excited np states of Na, K, Rb, and Cs. We note that for Li, the procedure of Eq. (7) was not used, but instead we employed the potential of Seitz[10] directly, i.e., without corrections. For comparison, the values of $E(np)$ and also $E(n_0s)$ as obtained from Moore's tables are presented in Table II.

The ratio involved is

$$\xi(np)=\frac{\delta E(np)}{|E_0(np)|}=\frac{a\,|E_{\text{exch}}(np)|}{|E_0(np)|}\ . \tag{9}$$

It is found that $|\xi(np)|$ is always less than 0.06. As an example, for K, $\xi(4p)=0.0227$, $\xi(5p)=0.0168$, and $\xi(6p)=0.0141$.

In Table III, we have given the expectation values $\langle r^{-3}\rangle$, $\langle r^{2}\rangle$, and $\langle r^{-1}\rangle$ over the valence np wave functions obtained in the present work. A comparison of the calculated $\langle r^{-3}\rangle$ values with those derived from the experimental values of the magnetic hyperfine structure and the fine structure will be made below (see Sec. III). For comparison with $\langle r^{-1}\rangle$, we have also listed in Table III the values of r_{max}, the radius of the outermost maximum of $v(np)$, and of the resulting $1/r_{\text{max}}$.

The perturbed wave functions $v_1'(nl\to l')$ obtained from Eq. (1), as well as the unperturbed functions $u_0'(nl)$, have been tabulated in a separate paper.[18] This paper also includes tables of the valence wave functions $v(np)$ which were determined from Eq. (7).

As mentioned above, for Li, we used the 1s wave function obtained by James.[9] This function is a hydrogenic function with effective nuclear charge $Z_0=2.69$. Thus,

$$u_0'(1s)=2Z_0^{3/2}re^{-Z_0r}\ . \tag{10}$$

The corresponding perturbation $v_1'(1s\to d)$ is given by[19]

$$v_1'(1s\to d)=\tfrac{1}{3}Z_0^{3/2}(1+\tfrac{1}{3}Z_0r)e^{-Z_0r}\ . \tag{11}$$

The factor $\frac{1}{3}Z_0^{3/2}$ equals 1.4706. The function $w_1'(1s\to d)$ listed in Table I of Ref. 18 is defined as $v_1'(1s\to d)(3/Z_0^{3/2})$, i.e., it goes to 1.00 at $r=0$. Thus we have

$$v_1'(1s\to d)=1.4706\,w_1'(1s\to d)\ . \tag{12}$$

TABLE I. Values of $E_{\text{exch}}(np)\equiv\int_0^\infty V_{\text{exch}}\,[v(np)]^2\,dr$, where V_{exch} is the Slater-type exchange corrected by the factor $\frac{2}{3}$ (Refs. 15 and 16). We have $n=n_0+i$, $i=0$, 1, and 2, and in the table $E_{\text{exch}}(np)$ is written as $E_{\text{exch}}(i)$. Note that $n_0=3$ for Na, $n_0=4$ for K, $n_0=5$ for Rb, and $n_0=6$ for Cs. The values of $E_{\text{exch}}(np)\equiv E_{\text{exch}}(i)$ are given in Rydberg units.

$-E_{\text{exch}}(np)$	Na	K	Rb	Cs
$-E_{\text{exch}}(0)$	0.051 49	0.065 72	0.069 36	0.076 48
$-E_{\text{exch}}(1)$	0.015 48	0.019 36	0.020 72	0.021 99
$-E_{\text{exch}}(2)$	0.006 747	0.008 460	0.009 207	0.009 835

TABLE II. Values of $|E(n_0s)|$ and $|E(np)|$ (in Rydberg units). For the np states, we have $n=n_0$, n_0+1, and n_0+2. The value of n_0 is listed in parentheses for each alkali (e.g., $n_0=3$ for Na). The values of $|E(nl)|$ were obtained from the tables of Moore (Ref. 2).

State	Li ($n_0=2$)	Na (3)	K (4)	Rb (5)	Cs (6)
n_0s	0.396 29	0.377 72	0.319 03	0.307 02	0.286 20
n_0p	0.260 48	0.223 10	0.200 35	0.190 94	0.180 97
$(n_0+1)p$	0.114 48	0.101 87	0.093 82	0.090 44	0.086 76
$(n_0+2)p$	0.063 95	0.058 39	0.054 72	0.053 15	0.051 42

The determination of the direct and exchange terms of R, namely, R_D and R_E, from the wave functions $v_1'(nl\to l')$ and $v_0(np)$ will now be outlined. The formalism is the same as that given in Ref. 5 (see pp. 11–13).

In the following discussion, the principal quantum number n of the external valence electron will be denoted by n_e, to distinguish it from the principal quantum number n of the core electron considered. The contribution to the direct term R_D due to the interaction of $(nl\to l')$ with the valence electron is given by

$$R_D(nl\to l'\,;\,n_ep)=\Gamma_D(nl\to l'\,;\,n_ep)/\langle r^{-3}\rangle_{n_ep}\ , \tag{13}$$

where $\Gamma_D(nl\to l'\,;\,n_ep)$ is defined by

$$\Gamma_D(nl\to l'\,;\,n_ep)=\int_0^\infty\frac{\gamma(nl\to l'\,;\,r)}{r^3}\,[v(n_ep)]^2\,dr, \tag{14}$$

TABLE III. Values of $\langle r^{-3}\rangle$, $\langle r^{2}\rangle$, and $\langle r^{-1}\rangle$ averaged over the alkali atom np wave functions. For comparison with $\langle r^{-1}\rangle$, the last two columns list the values of r_{max}, and $1/r_{\text{max}}$, where r_{max} is the radius of the outermost maximum of the np wave function. All values of $\langle r^n\rangle$ are in units of a_H^n.

State	$\langle r^{-3}\rangle$	$\langle r^{2}\rangle$	$\langle r^{-1}\rangle$	r_{max}	$1/r_{\text{max}}$
Li 2p	0.064 94	26.894	0.271 4	3.7	0.270 3
Li 3p	0.019 83	167.23	0.118 65	11.6	0.086 21
Li 4p	0.008 437	566.53	0.066 06	22.9	0.043 67
Na 3p	0.233 1	39.81	0.216 5	4.8	0.208 3
Na 4p	0.075 25	216.91	0.100 0	13.4	0.074 63
Na 5p	0.033 58	690.74	0.057 75	25.4	0.039 37
K 4p	0.414 49	52.42	0.185 13	5.7	0.175 4
K 5p	0.132 39	259.65	0.088 79	14.7	0.068 03
K 6p	0.059 12	790.35	0.052 51	27.4	0.036 50
Rb 5p	0.865 1	57.82	0.173 06	5.95	0.168 1
Rb 6p	0.286 2	280.58	0.084 43	15.3	0.065 36
Rb 7p	0.130 82	838.01	0.505 59	28.4	0.035 21
Cs 6p	1.416	67.54	0.158 99	6.7	0.149 3
Cs 7p	0.441 19	309.05	0.078 61	16.2	0.061 73
Cs 8p	0.202 3	901.32	0.047 79	29.2	0.034 25

and the function $\gamma(nl\rightarrow l';\, r)$ is given by

$$\gamma(nl\rightarrow l';\, r)=c(nl\rightarrow l')\left[\int_0^r u_0'(nl)v_1'(nl\rightarrow l')r'^2\,dr' + r^5\int_r^\infty u_0'(nl)v_1'(nl\rightarrow l')r'^{-3}\,dr'\right] . \quad (15)$$

The coefficients $c(nl\rightarrow l')$ have the following values: $\frac{8}{5}$ for $ns\rightarrow d$ and $nd\rightarrow s$; $\frac{48}{25}$ for $np\rightarrow p$; $\frac{72}{25}$ for $np\rightarrow f$; $\frac{16}{7}$ for $nd\rightarrow d$; and $\frac{144}{35}$ for $nd\rightarrow g$.

The expectation value $\langle r^{-3}\rangle_{n_e p}$ in the denominator of Eq. (13) is given by

$$\langle r^{-3}\rangle_{n_e p}=\int_0^\infty [v(n_e p)]^2 r^{-3}\,dr \, . \quad (16)$$

The values of $\langle r^{-3}\rangle_{n_e p}$ are listed in Table III. We note a slight change of notation from that of Ref. 5. Thus $n_e l_e$ is replaced by $n_e p$, and $w(n_e l_e)$ is changed to $v(n_e p)$ for the valence electron wave function.

For the exchange term due to the angular momentum L interaction between the perturbation $(nl\rightarrow l')$ and the valence electron $(n_e p)$, we have

$$R_E(nl\rightarrow l';\, n_e p;\, L)=\Gamma_E(nl\rightarrow l';\, n_e p;\, L)/\langle r^{-3}\rangle_{n_e p} \, , \quad (17)$$

where $\Gamma_E(nl\rightarrow l';\, n_e p;\, L)$ is defined by

$$\Gamma_E(nl\rightarrow l';\, n_e p;\, L)=-C(nl\rightarrow l';\, n_e p;\, L)\times K(nl\rightarrow l';\, n_e p;\, L) \, , \quad (18)$$

and the integral $K(nl\rightarrow l';\, n_e p;\, L)$ is given by

$$K(nl\rightarrow l';\, n_e p;\, L)=\int_0^\infty u_0' v(n_e p) g_L\, dr \, . \quad (19)$$

In Eq. (18), $C(nl\rightarrow l';\, n_e p;\, L)$ is an angular coefficient whose values for the cases of interest will be listed below. The function $g_L(r)$ in Eq. (19) is defined by

$$g_L(nl\rightarrow l';\, n_e p)=\frac{1}{r^{L+1}}\int_0^r v_1'(nl\rightarrow l')v(n_e p)r'^L\,dr' + r^L\int_r^\infty v_1'(nl\rightarrow l')v(n_e p)r'^{-L-1}\,dr' \, . \quad (20)$$

The values of $C(nl\rightarrow l';n_e p;\, L)$ are given in Table II of Ref. 5. For the cases of interest here, they are as follows:

$$C(ns\rightarrow d;\, n_e p;\, L=1)=\tfrac{4}{3},\quad C(np\rightarrow p;n_e p;\, L=2)=\tfrac{4}{25},$$
$$C(np\rightarrow f;n_e p;\, L=2)=\tfrac{36}{25},\quad C(nd\rightarrow d;\, n_e p;\, L=1)=\tfrac{4}{3},$$
$$C(nd\rightarrow d;n_e p;\, L=3)=\tfrac{12}{49},\quad C(nd\rightarrow g;\, n_e p;\, L=3)=\tfrac{72}{49}. \quad (20')$$

The L values are as indicated for each mode of excitation $nl\rightarrow l'$. We note that only for the case $nd\rightarrow d$ do we have two different L values contributing ($L=1$ and $L=3$). The total $R_E(nd\rightarrow d;\, n_e p)$ is the sum over the two L contributions, as explained in Ref. 5 [see Eq. (37)].

In view of Eqs. (19) and (20), $K(nl\rightarrow l';\, n_e p;\, L)$ is actually a double integral over the wave functions u_0', $v_1'(nl\rightarrow l')$, and $v(n_e p)$. The same is true of the integral for $\Gamma_D(nl\rightarrow l';\, n_e p)$ [see Eq. (14)]. These double integrals were evaluated by means of the computer program which has been described in Ref. 5 (see p. 15).

Because of the external nature of the valence np functions for the alkalis and the consequently small amount of overlap with the core wave functions, it had been previously expected that the exchange terms of R would be relatively small compared to the direct terms. This expectation was indeed borne out by the present calculations (except for Li). As an example, Table IV gives the terms $R_D(nl\rightarrow l';\, 6p)$ and $R_E(nl\rightarrow l';\, 6p)$ for the $6p$ state of Rb. The values of the ratio η are also listed, where $\eta(nl\rightarrow l';6p)$ is defined by

$$\eta(nl\rightarrow l';\, 6p)\equiv-\frac{R_E(nl\rightarrow l';\, 6p)}{R_D(nl\rightarrow l';\, 6p)} \, . \quad (21)$$

It is seen that except for the $1s\rightarrow d$ excitation, where $\eta=0.710$, η is indeed small in all cases ($\eta<0.25$), and for the combined radial and angular terms, we have $\eta_{\text{rad}}=0.0135$ and $\eta_{\text{ang}}=0.298$, respectively.

The resulting values of $R(nl\rightarrow l';n_e p)$ for the lowest three excited np states for the five alkalis are listed in Tables V–X. Note that all values of R have been multiplied by 10^2, except for Li (Table V), where no factor has been used, and for the angular terms for Cs (Table X), where the factor is 10^3.

In Tables VI–X, the last column gives the contributions $\gamma_\infty(nl\rightarrow l')$ to the total ionic antishielding

TABLE IV. Values of the direct and exchange contributions to $R(nl\rightarrow l';6p)$ for Rb $6p$. The direct and exchange terms are denoted by R_D and R_E, respectively. As indicated, these terms have been multiplied by 10^2. For each mode of excitation, the ratio η listed in the last column is defined by $\eta\equiv-R_E/R_D$. The radial terms are listed first, followed by the corresponding sums, denoted by Total (rad). The angular terms are listed in the bottom part of the table, followed by the corresponding sums: Total (ang). In the fourth column, $R=R_D+R_E$.

$nl\rightarrow l'$	10^2R_D	10^2R_E	10^2R	η
$2p\rightarrow p$	- 1.116	-0.017	- 1.133	-0.0152
$3p\rightarrow p$	- 4.189	~0	- 4.189	~0
$3d\rightarrow d$	- 1.299	+0.240	- 1.059	+0.185
$4p\rightarrow p$	-16.88	+0.094	-16.79	+0.0056
Total (rad)	-23.49	+0.317	-23.17	+0.0135
$1s\rightarrow d$	0.8840	-0.6275	0.2565	0.710
$2s\rightarrow d$	0.2816	-0.0279	0.2537	0.099
$2p\rightarrow f$	0.5401	-0.1228	0.4173	0.227
$3s\rightarrow d$	0.2150	-0.0215	0.1935	0.100
$3p\rightarrow f$	0.3607	-0.0535	0.3072	0.148
$3d\rightarrow g$	0.4363	-0.0413	0.3950	0.095
$4s\rightarrow d$	0.2458	-0.0579	0.1879	0.236
$4p\rightarrow f$	0.3996	-0.0491	0.3505	0.123
Total (ang)	3.3631	-1.0015	2.3616	0.298

TABLE V. Values of the terms of the shielding factor R for the $2p$, $3p$, and $4p$ excited states of lithium. R_D refers to the direct term, and R_E refers to the exchange term of R. The values of $\langle r^{-3}\rangle_{np}$ are in units a_H^{-3}.

R_D	0.1824	0.1738	0.1712
R_E	−0.0658	−0.0723	−0.0742
R(total)	0.1166	0.1015	0.0970
$1/(1-R)$	1.132	1.113	1.107
$\langle r^{-3}\rangle_{np}$(calc)	0.06494	0.01983	0.008437
$\langle r^{-3}\rangle_{np}$(exp)	0.06451	0.01909	...

factor[20] γ_∞. We have

$$\gamma_\infty(nl\to l')=c(nl\to l')\int_0^\infty u_0'(nl)v_1'(nl\to l')r^2\,dr \quad . \tag{22}$$

The last row of Tables VI–X gives the ratios ρ of the angular to the radial contributions for each case. Thus ρ is defined as

$$\rho_R\equiv-R_{\text{ang}}(n_ep)/R_{\text{rad}}(n_ep) \quad , \tag{23}$$

$$\rho_\gamma\equiv-\gamma_{\infty,\text{ang}}(n_ep)/\gamma_{\infty,\text{rad}}(n_ep) \quad , \tag{24}$$

pertaining to the atomic correction factor R and the ionic factor γ_∞, respectively. It is seen that the values of ρ_R and ρ_γ are always small compared to 1 and $\rho_\gamma<\rho_R$. Both ρ_γ and ρ_R decrease with increasing Z. Thus ρ_γ decreases from 0.113 for Na to 0.0191 for Cs. On the other hand, for a given atom, ρ_R increases somewhat with increasing n_e of the valence electron, e.g., from 0.0891 for Rb $5p$ to 0.1080 for Rb $7p$.

In connection with Tables VIII and X for Rb and Cs, we note that we have not listed the small contributions due to Rb $3d\to s$ and Cs $3d\to s$ and $4d\to s$. In order to estimate these terms, we note that the perturbations $v_1'(nd\to s)$ have been previously calculated[21] for the three following cases: $v_1'(3d\to s)$ for Cu$^+$; $v_1'(4d\to s)$ for Ag$^+$; and $v_1'(5d\to s)$ for Bi (see Sec. III of Ref. 21). The functions $v_1'(nd\to s)$ have the same general behavior and the same number of nodes as the corresponding perturbed functions $v_1'(nd\to d)$, although the $v_1'(nd\to s)$ are considerably smaller in magnitude and have opposite sign to $v_1'(nd\to d)$. In fact, by comparing the two types of functions in detail, it was found that $v_1'(nd\to s)/v_1'(nd\to d)$ is of the order of -0.1 throughout the range of r. Since the ratio of the angular coefficients $c(nl\to l')$ is

$$\frac{c(nd\to s)}{c(nd\to d)}=\frac{8/5}{16/7}=\frac{7}{10} \quad , \tag{25}$$

we expect that $\gamma_\infty(nd\to s)\sim-0.07\gamma_\infty(nd\to d)$. We

TABLE VI. Values of the contributions to R and to γ_∞ from the core excitations $nl\to l'$ for Na $3p$, $4p$, and $5p$. The values of $R(nl\to l'\,;n_ep)$ have been multiplied by 10^2, as indicated. In the rows labeled "total," "ang" refers to the angular perturbations $nl\to l'$ with $l'\neq l$; "rad" refers to the radial perturbations $nl\to l$. The last row lists the values of ρ defined as $\rho_R\equiv-R_{\text{ang}}(n_ep)/R_{\text{rad}}(n_ep)$ and $\rho_\gamma\equiv-\gamma_{\infty,\text{ang}}/\gamma_{\infty,\text{rad}}$.

$nl\to l'$	$10^2R(3p)$	$10^2R(4p)$	$10^2R(5p)$	$\gamma_\infty(nl\to l')$
$2p\to p$	−23.84	−19.02	−17.26	−5.389
$1s\to d$	1.657	1.611	1.598	0.0641
$2s\to d$	1.613	1.417	1.343	0.2286
$2p\to f$	2.515	2.239	2.141	0.3179
Total (ang)	5.785	5.267	5.082	0.6106
Total (ang + rad)	−18.05	−13.75	−12.18	−4.778
ρ_R or ρ_γ	0.243	0.277	0.294	0.113

TABLE VII. Values of the contributions to R and to γ_∞ from the core excitations $nl\to l'$ for K $4p$, $5p$ and $6p$. The values of $R(nl\to l';n_ep)$ have been multipled by 10^2, as indicated. In the rows labeled "total," "ang" refers to the angular perturbations $nl\to l'$ with $l'\neq l$; "rad" refers to the radial perturbations $nl\to l$. The last row lists the values of ρ defined as $\rho_R\equiv-R_{\text{ang}}(n_ep)/R_{\text{rad}}(n_ep)$ and $\rho_\gamma\equiv-\gamma_{\infty,\text{ang}}/\gamma_{\infty,\text{rad}}$.

$nl\to l'$	$10^2R(4p)$	$10^2R(5p)$	$10^2R(6p)$	$\gamma_\infty(nl\to l')$
$2p\to p$	−4.73	−4.41	−4.30	−1.217
$3p\to p$	−20.49	−16.60	−15.17	−17.14
$1s\to d$	0.655	0.645	0.644	0.0368
$2s\to d$	0.715	0.689	0.680	0.1016
$2p\to f$	1.075	1.040	1.029	0.1366
$3s\to d$	0.533	0.469	0.445	0.2979
$3p\to f$	0.963	0.852	0.813	0.4775
Total (rad)	−25.22	−21.01	−19.47	−18.36
Total (ang)	+ 3.94	+ 3.70	+ 3.61	+ 1.05
Total 10^2R or γ_∞	−21.28	−17.31	−15.86	−17.31
ρ_R or ρ_γ	0.156	0.176	0.185	0.0572

TABLE VIII. Values of the contributions to R and to γ_∞ from the core excitations $(nl\to l')$ for Rb $5p$, and $6p$. $7p$. The values of $R(nl\to l';n_ep)$ have been multiplied by 10^2, as indicated. In the rows labeled "total," "ang" refers to the angular perturbations $nl\to l'$ with $l'\neq l$; "rad" refers to the radial perturbations $nl\to l$. The last row lists the values of ρ defined as $\rho_R\equiv-R_{\text{ang}}(n_ep)/R_{\text{rad}}(n_ep)$ and $\rho_\gamma\equiv-\gamma_{\infty,\text{ang}}/\gamma_{\infty,\text{rad}}$.

$nl\to l'$	$10^2R(5p)$	$10^2R(6p)$	$10^2R(7p)$	$\gamma_\infty(nl\to l')$
$2p\to p$	−1.18	−1.13	−1.12	−0.45
$3p\to p$	−4.61	−4.19	−4.04	−3.93
$3d\to d$	−1.19	−1.06	−1.01	−1.23
$4p\to p$	−21.29	−16.79	−15.21	−43.8
$1s\to d$	0.258	0.257	0.256	0.0185
$2s\to d$	0.258	0.254	0.252	0.0444
$2p\to f$	0.423	0.417	0.415	0.0609
$3s\to d$	0.205	0.194	0.190	0.1033
$3p\to f$	0.325	0.307	0.301	0.1587
$3d\to g$	0.416	0.395	0.388	0.1926
$4s\to d$	0.221	0.188	0.176	0.3145
$4p\to f$	0.409	0.351	0.330	0.5522
Total (rad)	−28.27	−23.17	−21.38	−49.4
Total (ang)	+ 2.52	+ 2.36	+ 2.31	+ 1.45
Total 10^2R or γ_∞	−25.75	−20.81	−19.07	−48.0
ρ_R or ρ_γ	0.0891	0.1019	0.1080	0.0294

TABLE IX. Values of the contributions to R and to γ_∞ from the radial core excitations $nl \to l$ for Cs $6p$, $7p$, and $8p$. The values of $R(nl \to l; n_0 p)$ have been multiplied by 10^2, as indicated. The last four rows of the table give the sum over the radial terms [Total (rad)], the corresponding sum over the angular terms [Total (ang)] as obtained from the next table (see Table X), and the resulting total $10^2 R$ and γ_∞, as well as the ratios ρ.

$nl \to l$	$10^2R(6p)$	$10^2R(7p)$	$10^2R(8p)$	$\gamma_\infty(nl \to l)$
$2p \to p$	- 0.575	- 0.564	- 0.559	- 0.2669
$3p \leftarrow p$	- 1.57	- 1.50	- 1.47	- 1.660
$3d \to d$	- 0.369	- 0.355	- 0.349	- 0.3596
$4p \to p$	- 4.56	- 4.19	- 4.03	- 9.77
$4d \to d$	- 1.05	- 0.95	- 0.90	- 2.912
$5p \to p$	-17.77	-14.92	-13.59	-90.32
Total (rad)	-25.89	-22.48	-20.90	-105.29
Total (ang)	+ 1.90	+ 1.83	+ 1.80	+ 2.01
Total 10^2R or γ_∞	-23.99	-20.65	-19.10	-103.28
ρ_R or ρ_γ	0.0734	0.0814	0.0861	0.0191

thus obtain the following estimates: $\gamma_\infty(3d \to s) \cong (0.07)(1.23) \sim 0.09$ for Rb; and for Cs

$$\gamma_\infty(3d \to s) + \gamma_\infty(4d \to s) \cong 0.07(0.360 + 2.912) \cong 0.23 \ , \tag{26}$$

where we have used the values of $\gamma_\infty(3d \to d)$ and $\gamma_\infty(4d \to d)$ given in Tables VIII and IX. Thus the final value of $\gamma_{\infty,\mathrm{ang}}(\mathrm{Rb}^+)$ is 1.54, giving $\gamma_\infty(\mathrm{total}) = -47.9$. Similarly, the final value of $\gamma_{\infty,\mathrm{ang}}(\mathrm{Cs}^+)$ is 2.24, and hence $\gamma_\infty(\mathrm{total}) = -103.05$. It should, however, be noted that the above values of $\gamma_\infty(nd \to s)$ may be underestimates, and the actual values may conceivably be of the order of twice as large. In this case, $\gamma_{\infty,\mathrm{ang}}$ would be increased to 1.63 for Rb^+ and to 2.47 for Cs^+. In any case, it is seen that the correction to γ_∞ due to the $nd \to s$ modes is quite small for both Rb^+ and Cs^+.

In connection with the preceding discussion, it may be of interest to list the accurate values of $\gamma_{\infty,\mathrm{ang}}$ which have been obtained in Ref. 5 and in the present work: 0.248 for Li^+, 0.611 for Na^+, 1.050 for K^+, 1.464 for Cu^+, 1.54 for Rb^+, and 2.24 for Cs^+. These values show that $\gamma_{\infty,\mathrm{ang}}$ increases slowly with increasing Z, much more slowly than $|\gamma_{\infty,\mathrm{rad}}|$, and this accounts for the decrease of ρ_γ with Z, which has been pointed out above.

The contribution of the $(nd \to s)$ modes to R is even smaller than the contribution to γ_∞. In fact, the change of R for Rb^+ and Cs^+ is only ~ 0.001, as will now be shown. In order to estimate the terms $R(nd \to s)$, we note that the exchange integrals involved are the same as for $R_E(nd \to d)$, namely the $L = 1$ exchange interaction with the valence electron, with the same coefficient $(\frac{4}{5})$ in both cases. (There is also an $L = 3$ interaction for $nd \to d$, which, however, does not concern us here; see Table II of Ref. 5.) Thus we have

$$R_D(nd \to s) \cong -0.07 \, R_D(nd \to d) \ , \tag{27}$$

$$R_E(nd \to s) \cong -0.1 \, R_E(nd \to d; L = 1) \ , \tag{28}$$

where $R_E(nd \to d; L = 1)$ is the $L = 1$ term of the total $R_E(nd \to d)$. As an example, for Rb $6p$, where $R_D(3d \to d) = -1.299 \times 10^{-2}$, $R_E(3d \to d; L = 1) = +0.204 \times 10^{-2}$, we find

$$R(3d \to s; 6p) \cong [(0.07)(1.299) - (0.10)(0.204)] \times 10^{-2} = 0.705 \times 10^{-3} \ , \tag{29}$$

and similarly small values are obtained for Rb $5p$ (0.796×10^{-3}) and Rb $7p$ (0.675×10^{-3}). Of course, it should again be noted that these values may be underestimated, but even if they should have to be multiplied by a factor of 2–3, they would still represent a very small correction $(\lesssim 2 \times 10^{-3})$, presumably smaller than relativistic effects (which have not been included in the present work) or effects due to the inaccuracies of the core Hartree-Fock wave functions on the large terms of R, in particular, on $R(4p \to p)$ for the Rb.

For the case of Cs, a similar estimate using the values of $R_D(3d \to d)$, $R_E(3d \to d; L = 1)$, $R_D(4d \to d)$, and $R_E(4d \to d; L = 1)$ gives the following values for $R(3d \to s) + R(4d \to s)$: 0.947×10^{-3} for Cs $6p$, 0.864×10^{-3} for Cs $7p$, and 0.830×10^{-3} for Cs $8p$. In view of these results for both Rb and Cs, we have increased all values of R by $+0.001$, which is, of course, an essentially negligible correction, as discussed above.

The final values of R for the five alkali atoms are listed in Table XI. The bottom part of this table lists the resulting values of the correction factor $\mathfrak{C} \equiv 1/(1 - R)$ for the nuclear quadrupole moments. These factors will be used in Sec. III to obtain cor-

TABLE X. Values of the contributions to R and to γ_∞ from the angular core excitations $nl \to l'$ for Cs $6p$, $7p$, and $8p$. The values of $R(nl \to l'; n_0 p)$ have been multiplied by 10^3, as indicated. (Note the larger factor, as compared to 10^2 used in the preceding tables.) The last row labeled "Total (ang)" gives the sum over the angular contributions to $10^3 R$ and γ_∞, with the exception of the small $nd \to s$ terms which are estimated in the text.

$nl \to l'$	$10^3R(6p)$	$10^3R(7p)$	$10^3R(8p)$	$\gamma_\infty(nl \to l')$
$1s \to d$	1.597	1.593	1.591	0.01241
$2s \to d$	1.486	1.475	1.470	0.02790
$2p \to f$	2.600	2.586	2.578	0.03914
$3s \to d$	1.249	1.221	1.210	0.06837
$3p \to f$	1.679	1.645	1.631	0.08660
$3d \to g$	2.243	2.203	2.186	0.09964
$4s \to d$	1.294	1.231	1.203	0.1681
$4p \to f$	1.618	1.541	1.508	0.1998
$4d \to g$	2.106	2.003	1.957	0.2677
$5s \to d$	1.293	1.167	1.103	0.4568
$5p \to f$	1.836	1.645	1.558	0.5874
Total (ang)	19.001	18.310	17.995	2.0139

TABLE XI. Values of the quadrupole antishielding factors R and the corresponding correction factors $\mathcal{C} \equiv 1/(1-R)$. The n values of the states are written as $n=n_0$, n_0+1, and n_0+2, where n_0 is the principal quantum number of the ground state (n_0s), and is listed in parentheses for each alkali.

Alkali	Li ($n_0=2$)	Na (3)	K (4)	Rb (5)	Cs (6)
$R(n_0p)$	+0.117	-0.181	-0.213	-0.257	-0.239
$R(n_0+1,p)$	+0.102	-0.138	-0.173	-0.207	-0.206
$R(n_0+2,p)$	+0.097	-0.122	-0.159	-0.190	-0.190
$\mathcal{C}(n_0p)$	1.133	0.847	0.824	0.796	0.807
$\mathcal{C}(n_0+1,p)$	1.114	0.879	0.853	0.829	0.829
$\mathcal{C}(n_0+2,p)$	1.107	0.891	0.863	0.840	0.840

rected values of Q for a number of alkali isotopes.

The oscillator strengths of the $n_0s \to n_0p$ transition in the alkali atoms have been recently evaluated in a paper by Hameed, Herzenberg, and James.[22] These authors used Hartree-Fock-Slater wave functions which include the effect of exchange in an approximate manner, but do not include correlation effects of the valence electron with core electrons having antiparallel spin to that of the valence electron. Since our wave functions $v(n_0s)$ and $v(n_0p)$ include the effects of both exchange and correlation, they are expected to give more reliable values of $f_{n_0s \to n_0p}$ than those of Ref. 22. [We note that the alkali ground-state wave functions $v(n_0s)$ were obtained previously in connection with the calculation of the alkali atom dipole polarizabilities α_d. They are listed in Table I of Ref. 14, and also in Ref. 17.]

It is thus of interest to calculate the oscillator strengths using the wave functions $v(n_0s)$ and $v(n_0p)$. In fact, we can also calculate the corresponding oscillator strengths $f_{n_0s \to np}$ for $n=n_0+1$ and $n=n_0+2$, since we have the appropriate np wave functions.

From Eq. (4.2) of Ref. 22, $f_{n_0s \to np}$ is given by

$$f_{n_0s \to np} = (2m/3\hbar^2)(E_{np} - E_{n_0s}) \left| \int_0^\infty v_{n_0s} v_{np} r \, dr \right|^2 . \tag{30}$$

With the present normalization of v_{nl} [see Eq. (4)], and with E_{nl} in Rydberg units and distances in units a_H, Eq. (30) becomes

$$f_{n_0s \to np} = \tfrac{1}{3}(E_{np} - E_{n_0s})[(r)_i]^2 , \tag{31}$$

where $(r)_i$ is the matrix element (integral) of Eq. (30), with $i \equiv n - n_0$. The values of $(r)_i$ in units a_H are tabulated in Table XII, together with the energy differences $E_{np} - E_{n_0s}$ (in Rydberg units), denoted by ΔE_i, i.e.,

$$\Delta E_i \equiv E_{np} - E_{n_0s} \quad \text{for } n = n_0 + i . \tag{32}$$

In addition to $(r)_i$, ΔE_i, and the oscillator strengths $f_i \equiv f_{n_0s \to np} (i = n - n_0)$, Table XII also lists the matrix elements of r^{-2} between n_0s and np, i.e.,

$$(r^{-2})_{n_0s \to np} = \int_0^\infty v_{n_0s} v_{np} r^{-2} \, dr . \tag{33}$$

These matrix elements are presented because they were readily obtained from the computer program, and because the integral of Eq. (33) enters into the calculation of the core polarization effects on the f_i, as was pointed out by Hameed *et al.*[22]

The values of $f_{n_0s \to n_0p}$ (denoted by f_0 in Table XII) are generally in good agreement with the corresponding experimental values, as listed in Table 4 of Ref. 22. We will list here the calculated values of Table XII of the present paper, followed by the range of experimental values for each alkali: (i) Li calc: 0.753; expt: 0.72 ± 0.03; (ii) Na calc: 0.966; expt: 0.99 ± 0.06; (iii) K calc: 1.087; expt: 1.00 ± 0.06; (iv) Rb calc: 1.133; expt: 0.99 ± 0.06;

TABLE XII. Values of the matrix elements $(r)_{n_0s \to np}$ and $(r^{-2})_{n_0s \to np}$, and the energy differences $E_{np} - E_{n_0s} \equiv \Delta E$, and the resulting oscillator strengths $f_{n_0s \to np}$. If we write $n = n_0 + i (i = 0, 1,$ or $2)$, the matrix elements are denoted by $(r)_i$, $(r^{-2})_i$, and the ΔE by ΔE_i, the oscillator strengths by f_i. Note that $n_0 = 2$ for Li, $n_0 = 3$ for Na, $n_0 = 4$ for K, $n_0 = 5$ for Rb, and $n_0 = 6$ for Cs. Thus for Na, $f_1 = f_{3s \to 4p}$. $(r^n)_i$ is in units a_H^n; ΔE_i is in Rydberg units.

Quantity	Li	Na	K	Rb	Cs
$-(r)_0$	+4.078	4.329	5.241	5.411	5.851
$-(r)_1$	-0.2280	0.4169	0.4469	0.5932	0.6783
$-(r)_2$	-0.2026	0.1818	0.1758	0.2637	0.2933
$-(r^{-2})_0$	0.06405	0.07346	0.05197	0.04936	0.04302
$-(r^{-2})_1$	0.02261	0.03537	0.02482	0.02466	0.02099
$-(r^{-2})_2$	0.01265	0.02247	0.01583	0.01595	0.01366
ΔE_0	0.13581	0.15462	0.11868	0.11608	0.10523
ΔE_1	0.28181	0.27585	0.22521	0.21658	0.19944
ΔE_2	0.33234	0.31933	0.26431	0.25387	0.23478
f_0	0.7530	0.9659	1.0868	1.1328	1.2007
f_1	0.00488	0.01598	0.01499	0.02540	0.03059
f_2	0.00455	0.00352	0.00272	0.00588	0.00673

(v) Cs calc: 1.201; expt: 1.09 ± 0.10. The relevant experimental references are listed in Ref. 22 in connection with Table 4. We note that the agreement of our theoretical values with experiment is generally within one standard deviation, except for Rb, where the apparent disagreement is ~ 2 standard deviations. For Cs, we note there are two experimental determinations which differ appreciably, both as to the best experimental value and the magnitude of the experimental standard deviation. Thus Minkowski and Muhlenbruck[23] in 1930 obtained $f_0 = 0.980 \pm 0.10$, whereas the more recent experiment of Kvater and Meister[24] (in 1952) gave $f_0 = 1.188 \pm 0.012$. If the latter experimental value should be the more reliable (as indicated by the smaller assigned error), then our calculated value (1.201) would indeed be in very good agreement with experiment.

In connection with the present calculations of R, we note that the same method of solution of the inhomogeneous equation (1) (with r^{-3} replaced by r^k) has been used to calculate the dipole and quadrupole polarizabilities[3,6] α_d and α_q of several ions and atomic states, and also the shielding factors[25] σ_k for the $4f$ electrons in the rare-earth ions Pr^{3+} and Tm^{3+}.

III. DETERMINATION OF NUCLEAR QUADRUPOLE MOMENTS Q

The values of $\mathcal{C}$ calculated in Sec. II have been applied to obtain corrected values of the nuclear quadrupole moments Q for a number of alkali isotopes. In this connection, a recent review article of Fischer[26] was particularly useful.[27] Thus in Table 13 of Ref. 26, Fischer has tabulated the most accurate available values of the uncorrected quadrupole moments Q_{expt} for the nuclei Na^{23}, K^{40}, Rb^{87}, and Cs^{134}, as obtained from spectroscopic hyperfine measurements in several excited $np\ {}^2P_{3/2}$ states. The values of $\mathcal{C} = 1/(1-R)$ which were used in this table were those previously calculated by one of us (R. M. S), both in Ref. 4 and in further estimates. In the present work, we have recalculated the corrected values of Q, denoted by Q_{corr}, using the values of $\mathcal{C} = 1/(1-R)$ obtained in Sec. II. Thus

$$Q_{\mathrm{corr}} = Q_{\mathrm{expt}}\mathcal{C} = Q_{\mathrm{expt}}[1/(1-R)] \ . \tag{34}$$

The resulting values of Q_{corr}, together with the experimental values Q_{expt} are given in Table XIII. The experimental errors are indicated in parentheses (in units of 10^{-3}b).

The present values of Q_{corr} do not differ appreciably from those given by Fischer.[26] This is obviously due to the fact that the present values of $\mathcal{C}$ do not differ by a large amount from the estimates previously made, which were based on Ref. 4. However, it should be pointed out that the present values of $\mathcal{C}$ have a much smaller uncertainty (probably < 0.03) than the previous estimates, where the uncertainty[4] was ~ 0.15.

From the values of Table XIII, we have obtained the following weighted averages for the four nuclei Na^{23}, K^{40}, Rb^{87}, and Cs^{134}:

$$\begin{aligned} Q(\mathrm{Na}^{23}) &= +0.101 \pm 0.008 \text{ b}; \\ Q(\mathrm{K}^{40}) &= -0.067 \pm 0.008 \text{ b}; \\ Q(\mathrm{Rb}^{87}) &= +0.127 \pm 0.001 \text{ b}; \\ Q(\mathrm{Cs}^{134}) &= +0.364 \pm 0.002 \text{ b} \ . \end{aligned} \tag{35}$$

In obtaining these averages, we have used $1/\sigma_i^2$ for the weights, where σ_i is the standard deviation as listed in the last column of Table XIII (for Q_{corr}). The resulting standard deviation of the averages listed in Eq. (35) is given by $1/\sigma^2 = \sum_i (1/\sigma_i^2)$.

We will now obtain the corrected values of Q for the other alkali isotopes. For the case of K^{39}, the measurements of Schmieder, Lurio, and Happer[28] give $Q_{\mathrm{expt}} = 0.057 \pm 0.004$ b for the uncorrected value in the $5\,{}^2P_{3/2}$ state. Upon applying the appropriate correction factor, namely $\mathcal{C}_{5p} = 0.853$ (see Table XII), we obtain

$$Q_{\mathrm{corr}}(\mathrm{K}^{39}) = +0.049 \pm 0.004 \text{ b}. \tag{36}$$

Upon making use of the ratio

$$Q(\mathrm{K}^{39})/Q(\mathrm{K}^{41}) = 0.8215 \pm 0.0001 \tag{37}$$

obtained by Bonczyk and Hughes,[29] we find

$$Q_{\mathrm{corr}}(\mathrm{K}^{41}) = +0.060 \pm 0.005 \text{ b}. \tag{38}$$

Similarly, upon using the moment ratio

$$Q(\mathrm{Rb}^{85})/Q(\mathrm{Rb}^{87}) = 2.0669, \tag{39}$$

also obtained in Ref. 29, we find

$$Q_{\mathrm{corr}}(\mathrm{Rb}^{85}) = +0.263 \pm 0.002 \text{ b}. \tag{40}$$

We will now discuss the cesium isotopes Cs^{131}, Cs^{132}, Cs^{133}, Cs^{135}, and Cs^{137}. It should be noted at the outset that since the correction factors $\mathcal{C}_{6p}$, $\mathcal{C}_{7p}$, and $\mathcal{C}_{8p}$ for Cs differ only by small amounts from those used by zu Putlitz and co-workers,[30] the resulting corrected moments will not differ appreciably from those previously obtained.

We thus find for $\mathrm{Cs}^{131}\ 6p$

$$\begin{aligned} Q_{\mathrm{corr}}(\mathrm{Cs}^{131}\ 6p) &= -0.572(10) \times (0.807/0.791) \\ &= -0.583(10). \end{aligned} \tag{41}$$

Here $-0.572(10)$ b is the value obtained by Ackermann *et al.*,[30] and 0.807/0.791 is the ratio of the revised to the previous value of the correction factor $\mathcal{C}_{6p}$.

Similarly, upon correcting the value of Ref. 30 for $Q(\mathrm{Cs}\ 7p)$, we find

$$\begin{aligned} Q_{\mathrm{corr}}(\mathrm{Cs}^{131}\ 7p) &= -0.562(8) \times (0.829/0.817) \\ &= -0.570(8), \end{aligned} \tag{42}$$

TABLE XIII. Values of the experimental quadrupole moments $Q_{\rm exp}$ (without correction factor $\mathcal{C}$) and of the corresponding corrected moments $Q_{\rm corr}$ [see Eq. (34)]. The values of $\mathcal{C} = 1/(1-R)$ are listed in the fourth column of the table. $Q_{\rm exp}$ and $Q_{\rm corr}$ are in units of $b = 10^{-24}$ cm^2. The experimental values $Q_{\rm exp}$ were taken from the compilation of Fischer (Ref. 26).

Nucleus	State	$Q_{\rm exp}$	$1/(1-R)$	$Q_{\rm corr}$
Na^{23}	$3p\,^2P_{3/2}$	0.121(14)[a]	0.847	0.102(12)
Na^{23}	$4p\,^2P_{3/2}$	0.114(12)[a]	0.879	0.100(11)
K^{40}	$4p\,^2P_{3/2}$	-0.081(12)[b]	0.824	-0.067(10)
K^{40}	$5p\,^2P_{3/2}$	-0.079(14)[b]	0.853	-0.067(12)
Rb^{87}	$5p\,^2P_{3/2}$	0.160(1)[c]	0.796	0.127(1)
Rb^{87}	$6p\,^2P_{3/2}$	0.153(1)[d]	0.829	0.127(1)
Rb^{87}	$7p\,^2P_{3/2}$	0.147(2)[e]	0.840	0.123(2)
Rb^{87}	$8p\,^2P_{3/2}$	0.153(9)[f]	0.844	0.129(8)
Cs^{134}	$6p\,^2P_{3/2}$	0.455(3)[g]	0.807	0.367(3)
Cs^{134}	$7p\,^2P_{3/2}$	0.436(3)[h]	0.829	0.361(3)
Cs^{134}	$8p\,^2P_{3/2}$	0.427(8)[i]	0.840	0.359(7)

[a] D. Schönberner and D. Zimmermann, Z. Physik 216, 172 (1968).

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where 0.829/0.817 is the ratio of the corresponding factors $\mathcal{C}_{7p}$.

Finally, upon weighting the two results (41) and (42) according to the inverse-square standard deviations $1/\sigma_i^2$, we obtain the following final value:

$$Q(\mathrm{Cs}^{131}) = -0.575 \pm 0.006 \text{ b}. \tag{43}$$

The same procedure can be used for the values of Ackermann *et al.*[30] for $Q(\mathrm{Cs}^{132}\ 6p)$ and $Q(\mathrm{Cs}^{132}\ 7p)$. We find

$$Q_{\rm corr}(\mathrm{Cs}^{132}\ 6p) = +0.459(10)\times(0.807/0.791) = +0.468(10), \tag{44}$$

$$Q_{\rm corr}(\mathrm{Cs}^{132}\ 7p) = +0.467(30)\times(0.829/0.817) = +0.474(30). \tag{45}$$

The resulting weighted average is

$$Q(\mathrm{Cs}^{132}) = +0.469 \pm 0.010 \text{ b}. \tag{46}$$

In order to obtain the corrected value of $Q(\mathrm{Cs}^{133})$, we use the uncorrected value -0.0036 ± 0.0013 b, which was derived by Bucka *et al.*[31] from the earlier measurements of Althoff.[32] Since this result was obtained for the $7^2P_{3/2}$ state, we use the correction factor $\mathcal{C}_{7p} = 0.829$, and obtain

$$Q(\mathrm{Cs}^{133}) = (-0.0036 \pm 0.0013)\times 0.829 = -0.0030 \pm 0.0011 \text{ b}. \tag{47}$$

For Cs^{135} and Cs^{137}, we use the quadrupole coupling constants $B(\mathrm{Cs}^{135}) = 2.19 \pm 0.09$ Mc and $B(\mathrm{Cs}^{137}) = 2.23 \pm 0.09$ Mc, which were determined by Bucka, Kopfermann, and Otten.[31] By comparing these values with that for Cs^{134}, namely, $B(\mathrm{Cs}^{134}) = 18.07 \pm 0.12$ Mc (in the same $7^2P_{3/2}$ state), as obtained by Heinzelmann *et al.*,[33] we can deduce the corrected values of $Q(\mathrm{Cs}^{135})$ and $Q(\mathrm{Cs}^{137})$ from that of $Q(\mathrm{Cs}^{134}) = +0.364 \pm 0.002$ b [see Eq. (35)]. We thus obtain

$$Q(\mathrm{Cs}^{135}) = [(2.19 \pm 0.09)/18.07]\times 0.364 = +0.044 \pm 0.002 \text{ b}, \tag{48}$$

$$Q(\mathrm{Cs}^{137}) = [(2.23 \pm 0.09)/18.07]\times 0.364 = +0.045 \pm 0.002 \text{ b}. \tag{49}$$

For convenience, we have listed in Table XIV the 12 alkali isotope Q values which have been determined above [Eqs. (35)–(49)]. This table also includes the corrected values of $Q(\mathrm{Cu}^{63})$ and $Q(\mathrm{Cu}^{65})$. As mentioned above (in Sec. II), these values were previously obtained by Sternheimer[5] from a calculation of the correction factors $\mathcal{C}$ for the configurations $3d^94s^2$ and $3d^{10}4p$ of copper. The corresponding experimental values $Q_{\rm expt}$ were obtained by Fischer *et al.*[34] and by Ney,[35] respectively, for the two excited atomic states.

In connection with the calculations of the wave functions $v(np)$ from which the values of R are derived, it is of interest to compare the calculated $\langle r^{-3}\rangle_{np}$ values with those which have been derived for Rb np from the fine-structure splittings, by Feiertag and zu Putlitz.[36] Upon multiplying the entries for $\langle r^{-3}\rangle_{6W}$ in Table 3 of Ref. 36 by $(a_H/A)^3 = (0.5292)^3 = 0.1482$, we obtain $\langle r^{-3}\rangle_{5p} = 0.8006$, $\langle r^{-3}\rangle_{6p} = 0.2611$, $\langle r^{-3}\rangle_{7p} = 0.1183 a_H^{-3}$. The corresponding values calculated from the present wave functions are $\langle r^{-3}\rangle_{5p} = 0.8651$, $\langle r^{-3}\rangle_{6p} = 0.2862$, $\langle r^{-3}\rangle_{7p} = 0.1308 a_H^{-3}$ (see Table III). The agreement between the two sets of values is very good, with a maximum discrepancy of ~10% for $7p$. The good agreement which has been obtained here is presumably due to the inclusion of an effective correlation between the valence electron and the core by the use of the experimental energy eigenvalue $E_0(np)$ in the equation determining $v(np)$ [Eq. (7)]. The agreement to within ~10% may also be contrasted with the discrepancy which exists for Na $3p$ between the value of $\langle r^{-3}\rangle_{3p}$ obtained from the valence wave function of Fock and Petrashen[37] (see Ref. 4, Table I), and that obtained in the present work. The two values are $0.145 a_H^{-3}$, and $0.2331 a_H^{-3}$, respectively. Thus the Fock-Petrashen value which includes exchange effects but not correlation is too small by a factor of 1.61.

Besides the values of $\langle r^{-3}\rangle$ for Rb, we note that for Cs $7p$ and Cs $8p$, values of $\langle r^{-3}\rangle_{\text{expt}}$ have been obtained by Heinzelmann *et al.*[33] and by Knohl *et al.*,[38] respectively. In units of a_H^{-3}, these values are $\langle r^{-3}\rangle_{7p} = 0.3975$ and $\langle r^{-3}\rangle_{8p} = 0.1814$. For comparison, our calculated values are $\langle r^{-3}\rangle_{7p} = 0.4419$ and $\langle r^{-3}\rangle_{8p} = 0.2023$ (see Table III). As for Rb np, the agreement is quite close, with the discrepancy being < 12% in both cases.

TABLE XIV. Values of the corrected quadrupole moments Q_{corr} (in units of $b = 10^{-24}$ cm^2).

Nucleus	Q_{corr}
Na^{23}	+0.101 ± 0.008
K^{39}	+0.049 ± 0.004
K^{40}	-0.067 ± 0.008
K^{41}	+0.060 ± 0.005
Cu^{63}	-0.211 ± 0.004
Cu^{65}	-0.195 ± 0.004
Rb^{85}	+0.263 ± 0.002
Rb^{87}	+0.127 ± 0.001
Cs^{131}	-0.575 ± 0.006
Cs^{132}	+0.469 ± 0.010
Cs^{133}	-0.0030 ± 0.0011
Cs^{134}	+0.364 ± 0.002
Cs^{135}	+0.044 ± 0.002
Cs^{137}	+0.045 ± 0.002

The results of Table XIII give strong support to the assertion of zu Putlitz and co-workers[38,39] that the inclusion of the correction factor $1/(1-R)$ results in a better agreement between the values of Q_{corr} obtained from different atomic states, as compared to the poorer agreement among the uncorrected values Q_{expt}. This is particularly in evidence for the cases of Rb^{87} and Cs^{134}.

For the case of Rb, this point was made by Bucka *et al.*[39] by calculating

$$Q_{5p,\text{expt}}/Q_{7p,\text{expt}}(\text{Rb}^{85}, \text{Rb}^{87}) = 1.07 \pm 0.03, \qquad (50)$$

which they compared with the ratio $\mathcal{C}_{7p}/\mathcal{C}_{5p} = 0.845/0.787 = 1.074$, using the earlier values of $\mathcal{C}_{np}$, as derived from Ref. 4. With the present values of $\mathcal{C}_{np}$, we have

$$\mathcal{C}_{7p}/\mathcal{C}_{5p} = 0.840/0.796 = 1.055, \qquad (51)$$

which is also in essential agreement with the experimental result [Eq. (50)].

In a similar manner, for the case of Cs, Knohl *et al.*[38] have calculated

$$Q_{8p,\text{expt}}/Q_{7p,\text{expt}}(\text{Cs}^{134}) = 0.977 \pm 0.020, \qquad (52)$$

which they compared with the earlier value of the ratio $\mathcal{C}_{7p}/\mathcal{C}_{8p} = 0.817/0.832 = 0.982$. This ratio is practically unchanged, i.e., its present improved value is

$$\mathcal{C}_{7p}/\mathcal{C}_{8p} = 0.829/0.840 = 0.987, \qquad (53)$$

also in agreement with the experimental result, Eq. (52).

It can be concluded, in particular for Rb, that the slow increase of $\mathcal{C}_{np}$ with increasing n is verified experimentally, thereby giving additional support to the calculations of the quadrupole antishielding effect for atomic states.

In this connection, the experimental results for Cu $3d^9 4s^2$ and $3d^{10} 4p$, together with the calculations of Ref. 5 for copper, probably provide the strongest evidence for the atomic shielding and antishielding effects. Besides these two verifications (Rb and Cu), we may also note the earlier results of Murakawa[40] on the $5d$ and/or $6p$ states of La^{139}, Lu^{175}, and Hg^{201}, and the evidence obtained by Barnes, Mössbauer, Kankeleit, and Poindexter[41] for the existence of a weak shielding ($R \sim +0.2$) for the $4f$ electrons in the rare-earth ion Tm^{3+}. This value of R_{4f}, as well as Murakawa's result for $R_{5d} \sim -0.4$, have been confirmed theoretically[42] by Sternheimer in Refs. 5 (see Table VI) and 42 (see Table VII).

In this connection, we note that Childs and Goodman in a series of papers, have investigated the existence of quadrupole shielding and antishielding effects for atomic states. Thus, in connection with measurements[43] on the hyperfine structure in two configurations of V^{51} ($3d^3 4s^2$ and $3d^4 4s$), Childs[44] has concluded from the result

$$\frac{b_{3d}(d^3s^2)/b_{3d}(d^4s)}{a_{3d}(d^3s^2)/a_{3d}(d^4s)} = 1.00 \pm 0.03, \qquad (54)$$

that the shielding effects are nearly the same for both configurations. (Here a and b are, respectively, the magnetic moment and quadrupole moment coupling constants.)

On the other hand, for the case of Co^{59} which has been investigated for the two configurations $3d^7 4s^2$ and $3d^8 4s$, Childs and Goodman[45] found a differential shielding effect given by

$$\frac{Q(Co^{59},\ 3d^7 4s^2;\ {}^4F)}{Q(Co^{59};\ 3d^8 4s;\ {}^4F)} = \left(\frac{b_{3d}}{a_{3d}}\right)_{3d^7 4s^2} \Bigg/ \left(\frac{b_{3d}}{a_{3d}}\right)_{3d^8 4s} = 1.10 \pm 0.02\ , \qquad (55)$$

which indicates that the ratio $(1 - R_{3d^7 4s^2})/(1 - R_{3d^8 4s})$ is also given by Eq. (55), i.e., 1.10 ± 0.02. However, for Ni^{61}, a similar differential effect[46] was not observed, to within 3%.

We note that in a recent determination of $Q(La^{139})$ by Childs and Goodman,[47] the application of an antishielding correction factor $1/(1 - R_{5d})$, with[5] $R_{5d} \cong -0.3$, results in a value of $Q(La^{139})$, which is in good agreement with the earlier determination of Murakawa.[40]

In a recent paper, Childs[48] has investigated the hyperfine structure of 17 low-lying atomic levels of Tb^{159}, for both the configurations $4f^9 6s^2$ and $4f^8 5d 6s^2$. For the latter configuration, the magnetic dipole and quadrupole coupling constants (a_{nl} and b_{nl}) were obtained both for the $4f$ and $5d$ valence electrons. The situation is thus similar to that for Cu, where both the $3d$ and $4p$ coupling constants have been obtained.[5,34,35] In fact, it was found in Ref. 48 that, whereas

$$a_{4f}/a_{5d} = \langle r^{-3}\rangle_{4f}/\langle r^{-3}\rangle_{5d} = 2.96\ , \qquad (56)$$

the ratio

$$b_{4f}/b_{5d} = \langle r^{-3}\rangle_{4f}(1 - R_{4f})/\langle r^{-3}\rangle_{5d}(1 - R_{5d}) \qquad (57)$$

equals only 2.02, and is thus smaller than (56) by a factor

$$(1 - R_{5d})/(1 - R_{4f}) = 2.96/2.02 = 1.47\ . \qquad (58)$$

This value of 1.47 can thus be regarded as the experimental value of $(1 - R_{5d})/(1 - R_{4f})$.

On the other hand, upon using the values $R_{4f} = +0.10 \pm 0.05$ and $R_{5d} = -0.3$, which were estimated by one of us (R.M.S.) on the basis of the results of Refs. 5 and 42, we find the following theoretical value of the ratio:

$$(1 - R_{5d})/(1 - R_{4f}) = 1.3/(0.90 \pm 0.05) = 1.44 \pm 0.08. \qquad (59)$$

The close agreement of (58) and (59) is well within the limits of error of the theoretical and experimental determinations.

The magnitude of the effect $(1 - R_{5d})/(1 - R_{4f})$ is very similar to that previously found for Cu, in which case $(1 - R_{4p})/(1 - R_{3d}) = 1.429$. It is also noteworthy that if we denote by $n_1 l_1$ the values of nl pertaining to the R factor in the numerator, and by $n_2 l_2$ the values of nl in the denominator, the two atomic states are related by $n_2 = n_1 - 1$ and $l_2 = l_1 + 1$ in both cases, i.e., the state with the larger n and the smaller l value shows an antishielding effect, while the other state gives a shielding of Q.

IV. SUMMARY

In the present work, we have calculated the quadrupole antishielding factors R for 15 excited atomic states, namely the lowest three excited np states for each of the five alkali atoms. Special care was taken to obtain accurate np valence wave functions $v(np)$, by requiring that the effective potential should be so adjusted as to reproduce the observed atomic energy eigenvalue $E_0(np)$ for each state. The resulting wave functions $v(np)$ have been checked by calculating the oscillator strengths $f_{n_0 s \to np}$, where n_0 is the principal quantum number of the ground state. For $n = n_0$, the resulting calculated values are in good agreement with the corresponding experimental values of $f_{n_0 s \to n_0 p}$, as obtained by optical measurements. Furthermore, the expectation values $\langle r^{-3}\rangle_{np}$ which enter into the quadrupole hyperfine structure, are in good agreement with those derived from experiment by zu Putlitz and coworkers[33,36,38] for the cases of Rb and Cs.

The resulting values of R and $\mathcal{C} = 1/(1 - R)$ are generally close to the previous estimates made by one of us (R.M.S.) based on Ref. 4. However, the uncertainty in $\mathcal{C}$ is considerably reduced, from ~ 0.15 to ~ 0.03 for the present values. The application of the correction factors $\mathcal{C}$ to the values of Q_{exp} is shown in Table XIII. The final weighted averages of the corrected quadrupole moments are listed in Table XIV, which includes values for 12 alkali isotopes, and in addition, $Q(Cu^{63})$ and $Q(Cu^{65})$, which have been previously determined by the same procedure in Ref. 5.

ACKNOWLEDGMENTS

We wish to thank Professor G. zu Putlitz for suggesting a recalculation of the antishielding factors R for Rb and Cs, and for stimulating discussions. We are also indebted to Professor H. M. Foley and Professor A. J. Freeman for helpful discussions.

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*Work performed under the auspices of U.S. Atomic Energy Commission.

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