

model were carried out using the Thomas-Fermi expression (138). However, the values given by (139) differ from (138) by less than 12 percent (for $Z \geq 18$) in the region of importance near the nucleus (i.e., for $x \lesssim 2$). Thus at $x=2$, the curly bracket of (139) which replaces $(\chi x)^{\frac{1}{2}}$ in Eq. (138) has the value 0.779 for $Z=18$ and 0.732 for $Z=57$, as compared to $(\chi x)^{\frac{1}{2}}=0.698$.

These differences are even smaller for $x < 2$ and vanish as $x \rightarrow 0$. Hence the previous conclusions²⁰ about the Thomas-Fermi values of Δq are essentially unaffected by the inclusion of exchange in the statistical model.

I would like to thank Professor H. M. Foley for suggesting this problem and for several helpful and stimulating discussions.

Rotational Spectra of RbCl by the Molecular Beam Electric Resonance Method*

J. W. TRISCHKA† AND R. BRAUNSTEIN‡
Columbia University, New York, New York

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The rotational transitions for which $J=0 \rightarrow 1$ were studied by the molecular beam electric resonance method for the first three vibrational states, $v=0, 1, 2$, of $\text{Rb}^{85}\text{Cl}^{35}$ and the ground vibrational state, $v=0$, of $\text{Rb}^{87}\text{Cl}^{35}$. The molecular constants are:

	$\text{Rb}^{85}\text{Cl}^{35}$	$\text{Rb}^{87}\text{Cl}^{35}$
Y_{01} (Mc/sec)	2627.394 ± 0.002	2609.779 ± 0.003
$-Y_{11} \approx \alpha_e$ (Mc/sec)	13.601 ± 0.005	13.464 ± 0.005
$Y_{21} \approx \gamma_e$ (Mc/sec)	0.021 ± 0.002	0.021 ± 0.002
B_e (Mc/sec)	2627.414 ± 0.010	
r_e (Å)	2.78670 ± 0.00006	

The quadrupole (eqQ) and spin-rotation (c) interaction constants of $\text{Rb}^{85}\text{Cl}^{35}$ are:

	$v=0$	$v=1$	$v=2$
$(eqQ)_{\text{Rb}}$ (Mc/sec)	-52.675 ± 0.005	-52.306 ± 0.030	-51.903 ± 0.040
$(eqQ)_{\text{Cl}}$ (Mc/sec)	+ 0.774 ± 0.009	+ 0.612 ± 0.013	+ 0.470 ± 0.017
c_{Rb} (kc/sec)	+ 0.3 ± 0.3		
c_{Cl} (kc/sec)	0.0 ± 0.8		

For the $v=0$ state of $\text{Rb}^{87}\text{Cl}^{35}$, $(eqQ)_{\text{Rb}} = -25.485 \pm 0.006$ Mc/sec. The ratio of the Rb quadrupole moments is $Q_{85}/Q_{87} = 2.0669 \pm 0.0005$. The mass ratio of the Rb isotopes is $M_{85}/M_{87} = 0.9770163 \pm 0.0000045$.

I. INTRODUCTION

THROUGH their studies of KCl and KBr Carlson, Fabricand, Lee, and Rabi^{1,2} initiated an investigation by the molecular beam electric resonance method^{1,3} of the rotational spectra of the alkali halide molecules in their ground $^1\Sigma$ electronic states. We have continued this work by studying the $J=0 \rightarrow 1$ transitions of $\text{Rb}^{85}\text{Cl}^{35}$ in its three lowest vibrational states and of $\text{Rb}^{87}\text{Cl}^{35}$ in its ground vibrational state.

We selected RbCl in order to clarify further the nature of the Cl quadrupole interaction which shows unusual variations with vibration in KCl.² Also, at the outset of our work, the mass ratio of the Rb isotopes had not been determined with the accuracy made possible by the electric resonance method. Since then

Honig, Mandel, and Townes, using microwave absorption measurements of the rotational spectra of RbBr and RbI, have obtained this mass ratio with an accuracy equal to ours,⁴ while Collins, Johnson, and Nier have made mass spectrometric measurements which give a mass ratio of much greater accuracy.⁵

The hyperfine structure of the spectra yielded the Rb and Cl quadrupole and spin-rotation interaction constants, the ratio of the quadrupole moments of the Rb isotopes and the frequencies of the unperturbed rotational lines. The last-named frequencies gave the first three rotational constants of the molecules, the equilibrium internuclear distance and the mass ratio of the Rb isotopes. Of the above quantities the following have been measured by other methods: the magnitudes of the quadrupole and spin-rotation interactions of Rb^{87} in Rb^{87}Cl ,⁶ the ratio of the quadrupole moments of the Rb isotopes,⁷ the internuclear distance,⁸ and the mass ratio of the Rb isotopes.^{4,5} These constants, with the

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† On leave of absence from Syracuse University, Syracuse, New York, 1952-1953.

‡ Now at RCA Laboratories, Princeton, New Jersey.

¹ Lee, Fabricand, Carlson, and Rabi, Phys. Rev. **91**, 1395 (1953).

² Fabricand, Carlson, Lee, and Rabi, Phys. Rev. **91**, 1403 (1953).

³ H. K. Hughes, Phys. Rev. **72**, 614 (1947).

⁴ Honig, Mandel, and Townes (private communication).

⁵ Collins, Johnson, and Nier, Phys. Rev. **94**, 398 (1954).

⁶ D. I. Bolef and H. J. Zeiger, Phys. Rev. **85**, 799 (1952).

⁷ V. Hughes and L. Grabner, Phys. Rev. **79**, 314 (1950).

⁸ Maxwell, Hendricks, and Mosley, Phys. Rev. **52**, 968 (1937).

exception of the Rb spin-rotation interaction and Rb mass ratio, have been determined by us with much greater accuracy.

The apparatus and procedure used in our experiments has been described by others.^{1,2}

II. THEORY

The first order Hamiltonian used to explain and to make calculations from the hyperfine structure of the observed rotational spectra of RbCl is

$$\begin{aligned} \mathcal{H} = & -(eqQ)_{\text{Rb}} [3(\mathbf{I}_1 \cdot \mathbf{J})^2 + \frac{3}{2}(\mathbf{I}_1 \cdot \mathbf{J}) \\ & - \mathbf{I}_1^2 \mathbf{J}^2] / 2I_1(2I_1 - 1)(2J - 1)(2J + 3) \\ & - (eqQ)_{\text{Cl}} [3(\mathbf{I}_2 \cdot \mathbf{J})^2 + \frac{3}{2}(\mathbf{I}_2 \cdot \mathbf{J}) \\ & - \mathbf{I}_2^2 \mathbf{J}^2] / 2I_2(2I_2 - 1)(2J - 1)(2J + 3) \\ & + c_{\text{Rb}}(\mathbf{I}_1 \cdot \mathbf{J}) + c_{\text{Cl}}(\mathbf{I}_2 \cdot \mathbf{J}). \quad (1) \end{aligned}$$

The first two terms in Eq. (1) are the operators for the quadrupole interactions, while the last two terms are the operators for the spin-rotation interactions. The nuclear spin-spin interaction is negligible. The quadrupole interaction constant defined by Bardeen and Townes is eqQ .⁹ I_1 and I_2 , the Rb and Cl nuclear spins, have the values 5/2, 3/2, and 3/2 for Rb⁸⁵, Rb⁸⁷, and Cl³⁵, respectively. J is the rotational quantum number.

The observed spectral lines can be explained without the use of the last term in Eq. (1); i.e., $c_{\text{Cl}} = 0$ within experimental error. However, it is not permissible to omit this term from the Hamiltonian since it cannot be concluded that $c_{\text{Cl}} = 0$. Therefore, while only the first three terms in Eq. (1) were used to determine the interaction constants for these terms, all four terms were used to determine the errors in these constants and in c_{Cl} .

Because $(eqQ)_{\text{Rb}} \gg (eqQ)_{\text{Cl}}$, the energy levels obtained by the use of the first three terms in Eq. (1) are, in the notation of Bardeen and Townes,^{9,10}

$$\begin{aligned} W(F_1, F) = & E(F_1) + A(F_1, F_1) \\ & + \sum_{F_1' \neq F_1} [A(F_1, F_1')]^2 / [E(F_1) - E(F_1')] \\ & + (c_{\text{Rb}}/2)[F_1(F_1 + 1) - I_1(I_1 + 1) - J(J + 1)]. \quad (2) \end{aligned}$$

F_1 and F are defined by $\mathbf{F}_1 = \mathbf{I}_1 + \mathbf{J}$ and $\mathbf{F} = \mathbf{I}_2 + \mathbf{F}_1$. The first term in Eq. (2) is the first-order energy of the Rb quadrupole interaction, while the second and third terms are the first and second order energies of the Cl quadrupole interaction. The last term is contributed by the Rb spin-rotation interaction.

The Rb quadrupole interaction is large enough to produce an observable mixing of rotational states. When the matrix elements of Bardeen and Townes were used

to calculate this second order effect,¹¹ the first order energy levels of Rb⁸⁵Cl³⁵ were shifted by the following amounts: ($J=0, F_1=5/2$), -6.2 kc/sec; ($1, 3/2$), -0.6 kc/sec; ($1, 5/2$), -2.2 kc/sec; ($1, 7/2$), -3.0 kc/sec.

The rotational constants were calculated from equations most conveniently found in reference 2 and originally derived by Dunham.¹²

III. SPECTRA

The total beam, refocused beam, and line intensities were of the same order as those observed with KBr.² For the weakest line which could be observed, the decrease in refocused beam produced a change of 3×10^{-15} amp in the ion current. The corresponding change for the strongest line was 10×10^{-15} amp. The sensitivity of the dc amplifier, expressed in terms of galvanometer deflection, was 10^{-15} amp/cm.

The measured line width was usually 16 kc/sec, while the predicted width was 8 kc/sec.¹³ The discrepancy between the observed and predicted line widths suggests that the radiofrequency (rf) power was too high. However, when the rf power was reduced, it was usually found that the line intensity decreased with no corresponding decrease in line width. On the other hand, it was found for a few lines, observed under conditions for which the signal-to-noise ratio was exceptionally high, that when the rf power was reduced sufficiently the lines split symmetrically into two lines about 8 kc/sec wide, separated at their peaks by a frequency of 12 kc/sec. No satisfactory explanation of this effect has been found and further confirmation and study of it seems necessary. It is believed that this line splitting is a consequence of some structural feature of the apparatus and is not a feature of the spectra of RbCl. The position of a line for which this splitting was observed was taken as midway between the peaks of the two lines into which it was split.

All of the lines observed were for the transition $J=0 \rightarrow 1$. Since there is no hyperfine structure when $J=0$, the hyperfine structure of the spectrum for a given vibrational state is identical with that for the rotational state $J=1$ and is conveniently labeled with the (F, F_1) values for the levels of this state. The numbers of experimentally distinct lines predicted for one vibrational state are eleven for Rb⁸⁵Cl³⁵ and eight for Rb⁸⁷Cl³⁵.

Table I is a list of all of the lines observed for Rb⁸⁵Cl³⁵ and Rb⁸⁷Cl³⁵. The largest number of lines was observed for the ground vibrational state $v=0$ of Rb⁸⁵Cl³⁵ because these lines had the greatest intensities. The only missing line in this spectrum, the one for which $F=0$ and $F_1=3/2$, was too weak to observe. No effort was made to study the lines of Rb⁸⁵Cl³⁵ designated by $F_1=5/2$ and $v=1, 2$. The lines of the Rb⁸⁷Cl³⁵ spectrum specified by $F_1=3/2$ and $F=0, 2$ were too weak to

⁹ J. Bardeen and C. H. Townes, Phys. Rev. **73**, 97 (1948).

¹⁰ The first three terms in Eq. (2) are given in reference 1 in different notation. The last term comes from V. Hughes and L. Grabner, Phys. Rev. **79**, 829 (1950).

¹¹ J. Bardeen and C. H. Townes, Phys. Rev. **73**, 627 (1948).

¹² J. L. Dunham, Phys. Rev. **41**, 721 (1932).

¹³ H. C. Torrey, Phys. Rev. **59**, 293 (1941).

TABLE I. Frequencies in Mc/sec of observed lines for $J=0 \rightarrow 1$ transitions in RbCl. The numbers in parentheses designate the hyperfine structure levels (F, F_1) of the $J=1$ state.

Vibrational state	Rb ⁸⁵ Cl ³⁵	Rb ⁸⁷ Cl ³⁵
0	5248.596 (2, 3/2)	5212.474 (1, 2; 1/2)
	5248.566 (1, 3; 3/2)	
	5243.905 (4, 7/2)	5207.389 (2, 5/2)
	5243.855 (3, 7/2)	5207.332 (4, 5/2)
	5243.788 (5, 7/2)	
	5243.745 (2, 7/2)	
	5232.889 (1, 5/2)	5201.032 (1, 3; 3/2)
	5232.809 (4, 5/2)	
	5232.750 (2, 5/2)	
	5232.671 (3, 5/2)	
1	5221.421 (2, 3/2)	
	5221.395 (1, 3; 3/2)	
	5216.752 (4, 7/2)	
	5216.712 (3, 7/2)	
	5216.660 (5, 7/2)	
2	5216.630 (2, 7/2)	
	5194.306 (1, 3; 3/2)	
	5189.681 (4, 7/2)	
	5189.656 (3, 7/2)	
	5189.616 (5, 7/2)	
	5189.587 (2, 7/2)	

observe. No attempt was made to find all of the four lines of this spectrum for which $F_1=5/2$. The two lines of this group shown in the table were identified by their consistency with the other measured lines in the spectrum.

The remarks which follow constitute all that can be said with certainty about the relative intensities of the observed lines within a given vibrational state. The ratio of the intensities of the Rb⁸⁵Cl³⁵ lines designated by (1, 3; 3/2) and (2, 3/2) was 2:1. For the same molecule the lines for which $F_1=5/2$ were noticeably weaker than those for which $F_1=7/2$.

Since no spectra of RbCl³⁷ were observed, it is necessary to prove that the spectra found can in fact be ascribed to RbCl³⁵. Since the nuclear spins of Cl³⁵ and Cl³⁷ are the same, the hyperfine structure gives no clue to the identity of the Cl isotope, and the electron

TABLE II. Quadrupole and spin-rotation interaction constants of RbCl.

Quantity	Rb ⁸⁵ Cl ³⁵	Rb ⁸⁷ Cl ³⁵
$(eqQ)_{Rb}$ ($v=0$) (Mc/sec)	-52.675 ± 0.005	-25.485 ± 0.006
	-52.306 ± 0.030	
	-51.903 ± 0.040	
$(eqQ)_{Cl}$ ($v=0$) (Mc/sec)	$+ 0.774 \pm 0.009$	
	$+ 0.612 \pm 0.013$	
	$+ 0.470 \pm 0.017$	
c_{Rb} ($v=0$) (kc/sec)	$+ 0.3 \pm 0.3$	
c_{Cl} ($v=0$) (kc/sec)	0.0 ± 0.8	

diffraction value of the internuclear distance is not sufficiently reliable to act as a check on the present measurements. However, the calculated mass ratio of the Rb isotopes would have been significantly different from the value found from mass spectrometer measurements,⁵ which was not the case, had the spectra been those of RbCl³⁷.

The experimental probable error in the determinations of line positions was 2.0 kc/sec for all lines except the $v=2$ lines of Rb⁸⁵Cl³⁵. For these lines the error was 2.5 kc/sec.

IV. CALCULATIONS AND RESULTS

A least-squares analysis was made of the hyperfine structure lines to determine the constants given in Table II and the unperturbed rotational frequencies shown in Table III. Not only the magnitudes, but also the signs of the interaction constants in Table II were determined uniquely by the relative positions of the spectral lines. The errors in the two tables are probable errors.

In the calculations made from the $v=1,2$ spectra of Rb⁸⁵Cl³⁵ use was made of the values of c_{Rb} and c_{Cl}

TABLE III. Calculated frequencies of the unperturbed rotational transitions, $J=0 \rightarrow 1$, of RbCl.

Molecule	Freq. (Mc/sec)
Rb ⁸⁵ Cl ³⁵ ($v=0$)	5241.1919 ± 0.0006
	5214.074 ± 0.003
	5187.039 ± 0.004
Rb ⁸⁷ Cl ³⁵ ($v=0$)	5206.0987 ± 0.0012

determined from the $v=0$ spectrum of this molecule. When $(eqQ)_{Rb}$ was calculated from the Rb⁸⁷Cl³⁵ spectrum it was assumed that $(eqQ)_{Cl}$ and c_{Cl} had the values determined from the $v=0$ spectrum of Rb⁸⁵Cl³⁵ and that c_{Rb} for Rb⁸⁷ was equal to this constant for Rb⁸⁵ multiplied by the ratio of the nuclear g values of the two isotopes.

The quadrupole interactions of Rb and Cl for Rb⁸⁵Cl³⁵ may be summarized by empirical equations which show their variation with vibrational state. These equations are, for Rb⁸⁵ in Rb⁸⁵Cl³⁵,

$$(eqQ)_{Rb}(\text{Mc/sec}) = -52.675 + 0.38v; \quad (3)$$

and for Cl³⁵ in Rb⁸⁵Cl³⁵,

$$(eqQ)_{Cl}(\text{Mc/sec}) = 0.744 - 0.155v, \quad (4)$$

where v is the vibrational quantum number.

The ratio of the nuclear quadrupole moments of the Rb isotopes is, to the present accuracy, equal to the ratio of the quadrupole interactions for these isotopes, since the effect of the differences in zero point vibrations is negligible. From the data for $v=0$,

$$Q_{85}/Q_{87} = 2.0669 \pm 0.0005. \quad (5)$$

Calculations based on the rotational frequencies in Table III gave the rotational constants and the equilibrium internuclear distance listed in Table IV. The calculation of the Y 's in Table III required, in addition to the frequencies in Table II, the quantity $Y_{02} = -4B_e^3/\omega_e^2$. The vibrational constant, determined by Barrow and Caunt from the ultraviolet absorption spectra of RbCl,¹⁴ is 270 cm^{-1} . $Y_{02} = -1.1\text{ kc/sec}$ for Rb⁸⁵Cl³⁵. The information in Table III did not permit an independent calculation of the Y 's for Rb⁸⁷Cl³⁵. Therefore, use was made of the well established relationships between the Y 's of different isotopic species.¹² Within experimental error γ_e for Rb⁸⁷Cl³⁵ is the same as γ_e for Rb⁸⁵Cl³⁵, while $\alpha_e^{87} = (\mu_{85}/\mu_{87})^{3/2}\alpha_e^{85}$, where μ is the reduced mass of the molecule. The calculation of Y_{01} for Rb⁸⁷Cl³⁵ then required these values of γ_e and α_e , the above value of Y_{02} , and the rotational frequency for $v=0$ in Table III.

The quantity B_e differs from Y_{01} by a small correction term¹ which involves all of the Y 's in Table III and the vibrational constants ω_e and $\omega_e x_e$.

$$B_e(\text{Mc/sec}) = Y_{01} + (0.020 \pm 0.010), \quad (6)$$

where the error is almost entirely the result of the errors in ω_e and $\omega_e x_e$. For the calculation of r_e from B_e the additional quantities required were the masses^{5,15} of Rb and Cl and the fundamental constants h/e and F .¹⁶ The error in r_e is entirely the result of the errors in h/e and F .

Within the present experimental accuracy the ratio of the reduced masses of Rb⁸⁵Cl³⁵ and Rb⁸⁷Cl³⁵ is

$$\mu_{85}/\mu_{87} = (Y_{01})_{87}/(Y_{01})_{85}. \quad (7)$$

When Eq. (7) was used in conjunction with the formula^{1,2} for the atomic mass ratio, the mass ratio of the Rb isotopes was found to be

$$M_{85}/M_{87} = 0.9770163 \pm 0.0000045. \quad (8)$$

V. DISCUSSION

Bolef and Zeiger,⁶ using the molecular beam magnetic resonance method at zero field, found $|eqQ|_{87}(\text{Mc/sec}) = 25.37 \pm 0.04$ for the Rb quadrupole interaction in the ground vibrational state of Rb⁸⁷Cl. This disagrees by more than the experimental errors with the present value of $25.485 \pm 0.004\text{ Mc/sec}$. If the source of this disagreement is sought in the present experiments, it may be suggested that the observed lines for Rb⁸⁷Cl³⁵ may not be properly identified since measurements were not made on all of the eight lines predicted. However, any different identification of these lines makes them inconsistent among themselves by more than their experimental errors and also causes a

¹⁴ R. F. Barrow and A. D. Caunt, Proc. Roy. Soc. (London) **A219**, 120 (1953).

¹⁵ Collins, Nier, and Johnson, Phys. Rev. **84**, 717 (1951).

¹⁶ J. W. M. DuMond and E. R. Cohen, Revs. Modern Phys. **25**, 691 (1953).

TABLE IV. Molecular constants of RbCl.

Constant	Rb ⁸⁵ Cl ³⁵	Rb ⁸⁷ Cl ³⁵
$Y_{01}(\text{Mc/sec})$	2627.394 ± 0.002	2609.779 ± 0.003
$-Y_{11} \approx \alpha_e(\text{Mc/sec})$	13.601 ± 0.005	13.464 ± 0.005
$Y_{21} \approx \gamma_e(\text{Mc/sec})$	0.021 ± 0.002	0.021 ± 0.002
$B_e(\text{Mc/sec})$	2627.414 ± 0.010	
$r_e = 2.78670 \pm 0.00006\text{ \AA}$		

serious disagreement between the mass ratio of the Rb isotopes determined from the present experiments and the ratio calculated from the mass spectrometric measurements. In Table V the quadrupole constants obtained by the zero field magnetic resonance method and by the electric resonance method are compared. Although all of the differences shown in the last column of the table have the same sign, only the difference for RbCl is greater than the experimental errors.

In the presentation of their results Bolef and Zeiger use the equation

$$eqQ = eqQ + eq^{(v)}Qv + eq^{(J)}QJ(J+1). \quad (9)$$

They find $eq^{(v)}Q/h = -0.230 \pm 0.050\text{ Mc/sec}$ for Rb⁸⁷ in Rb⁸⁷Cl. When the coefficient of the second term in Eq. (3) of the present paper is multiplied by the ratio of the Rb quadrupole interactions a value of $0.18 \pm 0.03\text{ Mc/sec}$ is obtained for $eq^{(v)}Q/h$ for Rb⁸⁷. It is seen that the two values agree within experimental error. Bolef and Zeiger found that eqQ and $eq^{(v)}Q$ have opposite signs, in agreement with the present results shown in Eq. (3).

The magnitude of the spin-rotation constant for Rb obtained in the present experiments agrees within experimental error with the more accurate value for Rb⁸⁷, $1.0 \pm 0.1\text{ kc/sec}$, found by Bolef and Zeiger who could not determine the sign of this constant.

The ratio of the quadrupole moments of the Rb isotopes found by Hughes and Grabner for RbF, 2.07 ± 0.01 ,⁷ agrees within the range of experimental errors with the more accurate value found in the present experiments.

The mass ratio of the Rb isotopes, 0.9770191 ± 0.0000022 , computed from the masses given by Collins, Johnson, and Nier⁵ agrees within experimental error with the less accurate value obtained in the present experiments. Possible difficulties in the in-

TABLE V. Comparison of quadrupole interaction constants determined by the magnetic resonance method at zero field and by the electric resonance method. The constants are given for the ground vibrational state.

Constant	Magnetic (Mc/sec)	Electric (Mc/sec)	Difference (El.-Mag.)
$ eqQ _{01}$ of TICl ³⁵	15.788 ± 0.020	15.795 ± 0.004	$+0.007$
$ eqQ _{01}$ of TICl ³⁷	12.425 ± 0.020	12.446 ± 0.003	$+0.021$
$ eqQ _{\text{Rb}}$ of Rb ⁸⁷ F	33.96 ± 0.02	34.00 ± 0.06	$+0.04$
$ eqQ _{\text{Rb}}$ of Rb ⁸⁷ Cl	25.37 ± 0.04	25.485 ± 0.004	$+0.115$

TABLE VI. Comparison of the alkali quadrupole interactions in the alkali halides. The interactions in the alkali fluorides are taken as standards for each row.

Alkali	F	Halogen		
		Cl	Br	I
Li	1.00	0.47	0.45	0.42
Na	1.00	0.67	0.57	0.48
K	1.00	0.71	0.63	—
Rb	1.00	0.75	—	—

terpretation of the mass ratio computed from the molecular constants of the alkali halides are discussed in reference 1.

The most remarkable feature of the quadrupole interactions in RbCl is the positive sign of the Cl^{35} interaction. Since the quadrupole moment of Cl^{35} is negative, q_{Cl} , the electric field gradient at the Cl nucleus, must be negative in RbCl. This sign of q is the opposite of that found experimentally for q at the halogen nuclei of KCl and KBr;^{1,2} it is also the opposite of the sign predicted by the theory of Inglis¹⁷ and the theory of Foley, Sternheimer, and Tycko.¹⁸ The antishielding effect, which is the central effect considered by the latter authors, is apparently so submerged by other effects in RbCl that a reversal in the sign of q results. Finally, the negative sign of q in RbCl contradicts the empirically based conjecture made by Logan, Cote, and Kusch that q at each nucleus is positive in every diatomic molecule.¹⁹

It is of interest to note that the second term in Eq. (4), giving the variation of the Cl interaction con-

stant with vibration in RbCl, has not only the same algebraic sign as it does in KCl,¹ but also nearly the same magnitude.

The sign of the Rb quadrupole interaction in RbCl has been determined for the first time in the present experiments. That this sign is the same as that found for the Rb interaction in RbF gives further support to the suggestion made by Logan, Cote, and Kusch that the signs of the alkali interactions are the same in all alkali halides having a common alkali atom.

Some interesting regularities in the magnitudes of the alkali quadrupole interactions in the alkali halides can be shown with the aid of Table VI. Each number in the table is the ratio of the alkali quadrupole interaction in a given alkali halide to the interaction in the fluoride of the same alkali. The alkali halide for which each ratio is given is specified by the row and column headings for that ratio.²⁰ An examination of Table VI shows that the ratios decrease monotonically from left to right and increase monotonically from top to bottom.

VI. ACKNOWLEDGMENTS

We wish to express our thanks to Professor I. I. Rabi and Professor P. Kusch for the opportunity to use the electric resonance apparatus at Columbia University and for their aid and advice while the present work was being performed. We are also indebted to Dr. Charles Lee for many helpful discussions during the performance of this work.

²⁰ The values of the quadrupole interactions in RbCl, KCl, and KBr come from the present paper and from references 1 and 2, respectively. All other values are tabulated in reference 19 where references to the original papers are given.

¹⁷ D. R. Inglis, *Revs. Modern Phys.* **25**, 390 (1953).

¹⁸ Foley, Sternheimer, and Tycko, *Phys. Rev.* **93**, 734 (1954).

¹⁹ Logan, Cote, and Kusch, *Phys. Rev.* **86**, 280 (1952).