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# University of California

## Ernest O. Lawrence Radiation Laboratory

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## I. INTRODUCTION

Over the past years, the molecular beam electric resonance technique of radio-frequency spectroscopy<sup>1,2</sup> has become established as a very accurate method for determining molecular dipole moments and studying interactions between nuclear moments and the molecular fields. The work described here forms part of a series of investigations on the radio-frequency spectra of the alkali halides. More accurate values of dipole moments and quadrupole coupling constants provide the test for a polarizable ion model for these molecules.<sup>3</sup>

The radio-frequency Stark spectra of Na<sup>23</sup>Cl<sup>35</sup> has been studied by Gold.<sup>4</sup> He obtained values of dipole moments for the first three vibrational states of the molecules. The quality of the spectra, however, did not permit an accurate determination of the hyperfine interaction constants. An approximate value of 5.6 MHz was given for the quadrupole coupling constant (eqQ) of both the Na<sup>23</sup> and Cl<sup>35</sup> nucleus.

A number of studies have been conducted to determine hyperfine interactions in Na<sup>23</sup>Cl<sup>35</sup> by the molecular beam magnetic resonance (MBMR) method.<sup>5,6,7,8</sup> The widely different values of the Na<sup>23</sup> coupling constant in Na<sup>23</sup>Cl<sup>35</sup>, however, lead one to doubt the accuracy of the method.

Up until the present work, the spectra of the isotopic molecule Na<sup>23</sup>Cl<sup>37</sup> have not been studied. The radio-frequency Stark spectra of Na<sup>23</sup>Br<sup>79</sup> and Na<sup>23</sup>Br<sup>81</sup> have not been studied before. Microwave measurements<sup>9</sup> gave a value of  $58 \pm 2$  MHz for (eqQ)<sub>Br79</sub> and magnetic (MBMR) measurements<sup>7,10</sup> indicate a value of 4.7 to 4.9 MHz for (eqQ)<sub>Na23</sub> in Na<sup>23</sup>Br<sup>79</sup>.

The incorporation of a signal averaging system to the molecular beam spectrometer in this laboratory has permitted the re-examination of the radio-frequency spectra of these molecules. From an analysis of the hyperfine

components of the Stark spectra, more accurate values of dipole moments and  $(eqQ)$ 's have been obtained as a function of vibrational state. An assignment of the  $(eqQ)$ 's in  $\text{Na}^{23}\text{Cl}^{35}$  to the respective nuclei was made possible after a determination of  $(eqQ)_{\text{Cl}^{37}}$  from the  $\text{Na}^{23}\text{Cl}^{37}$  spectra. Values of dipole moments of the two NaBr isotopes have been measured for the first time together with the  $(eqQ)$  of the various nuclei.

## II. EXPERIMENTAL APPARATUS AND PROCEDURES

### A. Molecular Beam Spectrometer

The experimental apparatus used in this work is that built by Hebert<sup>11</sup> and has been used in a number of previous investigations carried out in this laboratory.<sup>3,12</sup> Since it has been described in detail before, only a brief summary will be given here.

The spectrometer consists essentially of four high vacuum chambers which house an oven, a state-selecting inhomogeneous electric field (A-field), an extremely homogeneous electric field (C-field), a refocusing inhomogeneous electric field (B-field), a surface ionization detector, a  $60^\circ$  sector magnet mass spectrometer and a 14-stage electron multiplier. A schematic diagram of the apparatus is shown in Fig. 1.

The source ovens used in these experiments were  $3/8$ " diam  $\times$  0.010" wall  $\times$  6-1/2" long tubes of Platinum-Iridium alloy (80%-20%), with 0.25" long by 0.005" wide slit at the middle. Sample is placed in the oven and heated to about  $850^\circ\text{C}$  for NaCl and  $800^\circ\text{C}$  for NaBr by resistance heating. Sample effuses from the source to a buffer chamber and on to a main chamber which houses the deflecting fields and the detector. The inhomogeneous A and B fields are of the

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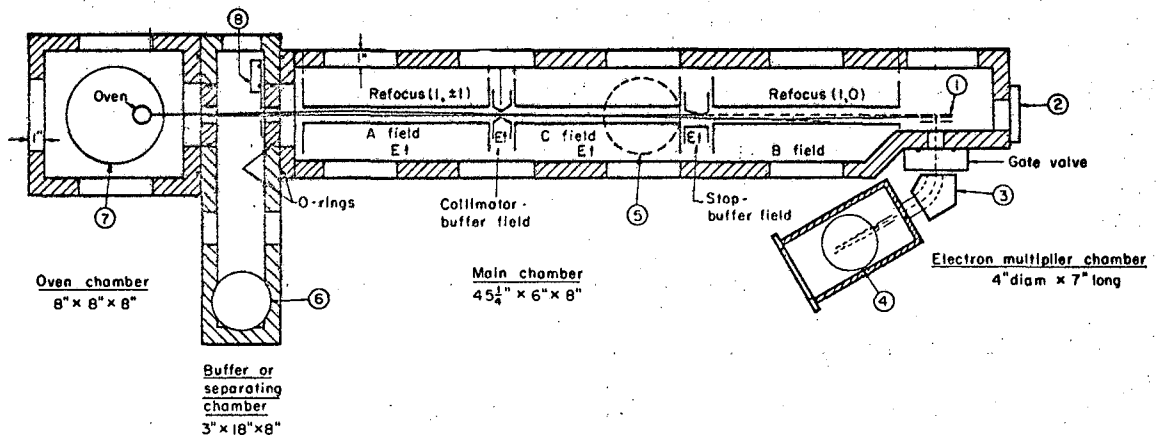
ABSTRACT

The radio-frequency Stark spectra of Na<sup>23</sup>Cl<sup>35</sup>, Na<sup>23</sup>Cl<sup>37</sup>, Na<sup>23</sup>Br<sup>79</sup> and Na<sup>23</sup>Br<sup>81</sup> have been observed by the molecular beam electric resonance method of molecular spectroscopy. Analysis of spectral lines obtained by observing transitions of the type  $J = 1, M_J = \pm 1 \rightarrow J = 1, M_J = 0$  at strong electric fields yielded molecular dipole moments and nuclear hyperfine interaction constants for the different vibrational states of the molecules.

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





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- |                 |               |                 |
|-----------------|---------------|-----------------|
| $l_0 = 12.3$ cm | $l_4 = 7$ cm  | $l_A = 30.5$ cm |
| $l_1 = 20.6$ cm | $l_5 = 55$ cm | $l_B = 30.5$ cm |
| $l_2 = 4$ cm    | $l_6 = 69$ cm | $l_C = 25.4$ cm |
| $l_3 = 5$ cm    |               |                 |

Fig. 1. Schematic diagram (top view) of the electric resonance apparatus. Field lengths and chambers are to scale. Field gaps and beam displacements are exaggerated. Unshaded areas in chamber walls represent access ports. (Dimensions refer to internal chamber sizes.)

- (1) Hot wire and ion accelerator
- (2) Glass port cover for optical alignment
- (3) Permanent magnet, 60°, 1-cm gap
- (4) Outlet, 4 in. diam, to liquid nitrogen trap and oil-diffusion pump
- (5) Outlet, 5 in. diam, to liquid nitrogen trap and oil-diffusion pump
- (6) Outlet, 4 in. diam, to gate valve, liquid nitrogen trap, and oil-diffusion pump
- (7) Outlet, 6 in. diam, to gate valve, liquid nitrogen trap, and oil-diffusion pump
- (8) Gate valve and beam flag.

dipole type, 30.5 cm long, and permit the selection of a certain  $(J, M_J)$  quantum state and refocusing of the same state ("flop-out" experiments) or another ("flop-in" experiment).<sup>1</sup>  $J$  is the rotational angular momentum quantum number for the molecule and  $M_J$  is its component in the direction of the electric field. The homogeneous Stark field (C-field) consists of 2 parallel plate electrodes 25.4 cm long and separated by  $1.000001 \pm 0.000001$  cm at 20°C. Field homogeneity is within 7 ppm. Field measurement is made via a Guildline ratio box with a Leeds and Northrup Guarded Potentiometer calibrated with a bank of unsaturated standard cells. Uncertainty in the C-field voltage measured is about 2 parts in 10.<sup>5</sup> Radiofrequency voltage is introduced in the C-field region in a manner first introduced by Trischka.<sup>13</sup> The ribbon beam is detected by surface ionization on a 0.010" wide  $\times$  0.001" thick Tungsten filament heated to about 1650°K. The ions evaporating from the hot filament are accelerated, mass analyzed using a 60° sector magnet and focussed on a 14-stage electron multiplier. The ion current is then amplified to drive a chart recorder for visual display of the spectra or the ion pulses from the multiplier amplified and counted for a digitalized output.

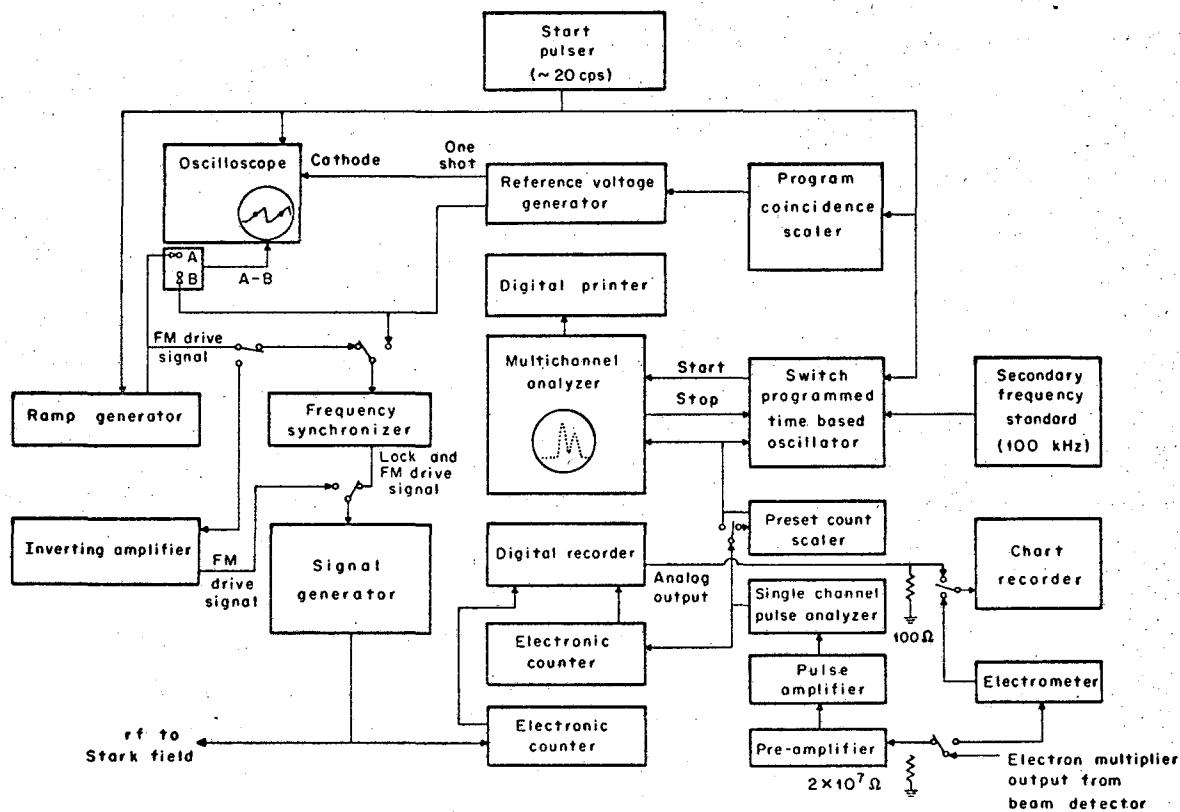
#### B. Signal Averaging System

Spectra of sufficient quality to enable the accurate determination of dipole moments and coupling constants for the sodium halides were made possible only by the incorporation of a signal averaging system for the recovery of weak signals buried in noise. The resonance signals from both NaCl and NaBr molecules are considerably weaker compared to that of the lighter alkali halides. This is due to an intrinsic reduction in beam intensity brought about by the size or

mass of the molecule<sup>14</sup> and the large residual noise level due to Na impurities evaporating from the hot wire detector. With NaCl, line measurements obtained without using the signal averaging system have at best a signal to noise ratio of 3:1; for NaBr, about 1:1. With signal averaging, considerable enhancement of the signal to noise ratio is obtained.

A block diagram of the signal averaging system used is shown in Fig. 2. This system will be described in greater detail elsewhere<sup>15</sup> and only a brief description will be given here. A list of the electronic equipment used is given in Table A-I of the Appendix. The principle of the method is based on the fact that in counting statistics, the fluctuation about the average number of counts is equal to the square root of the number of counts, for one standard deviation. The signal, on the other hand, grows in time in direct proportion to the number of counts accumulated. Thus if one could repeatedly scan the region of the spectrum in which a resonance line appears, store and average the signal intensities over long periods, one in effect obtains an enhancement of the signal to noise ratio over that obtainable by just a single sweep.

The system shown is used to produce a linear frequency or voltage sweep to scan the spectral region of interest several times a second. Basically, a synchronizer (Hewlett Packard 8708A) is used to phase lock the frequency of the signal generator (Hewlett Packard 608F) and a ramp voltage of variable frequency and amplitude used to sweep over a certain frequency range by modulating the signal generator view the synchronizer. With this mode of operation, sweep widths of a few hundred KHz were obtained. In exact step with this repetitive frequency sweep, is an "intensity counts" sweep provided by a switch programmed time base oscillator. This "intensity vs. time" sweep is designed to store and accumulate the count information in a multi-channel analyzer (Norther Scientific NS-610).



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Fig. 2. Block diagram of signal averaging system.

A triggering signal from the pulse generator (Electronics Inc., Model 301) starts both the frequency and multi-channel analyzer sweeps and ensures their synchronization.

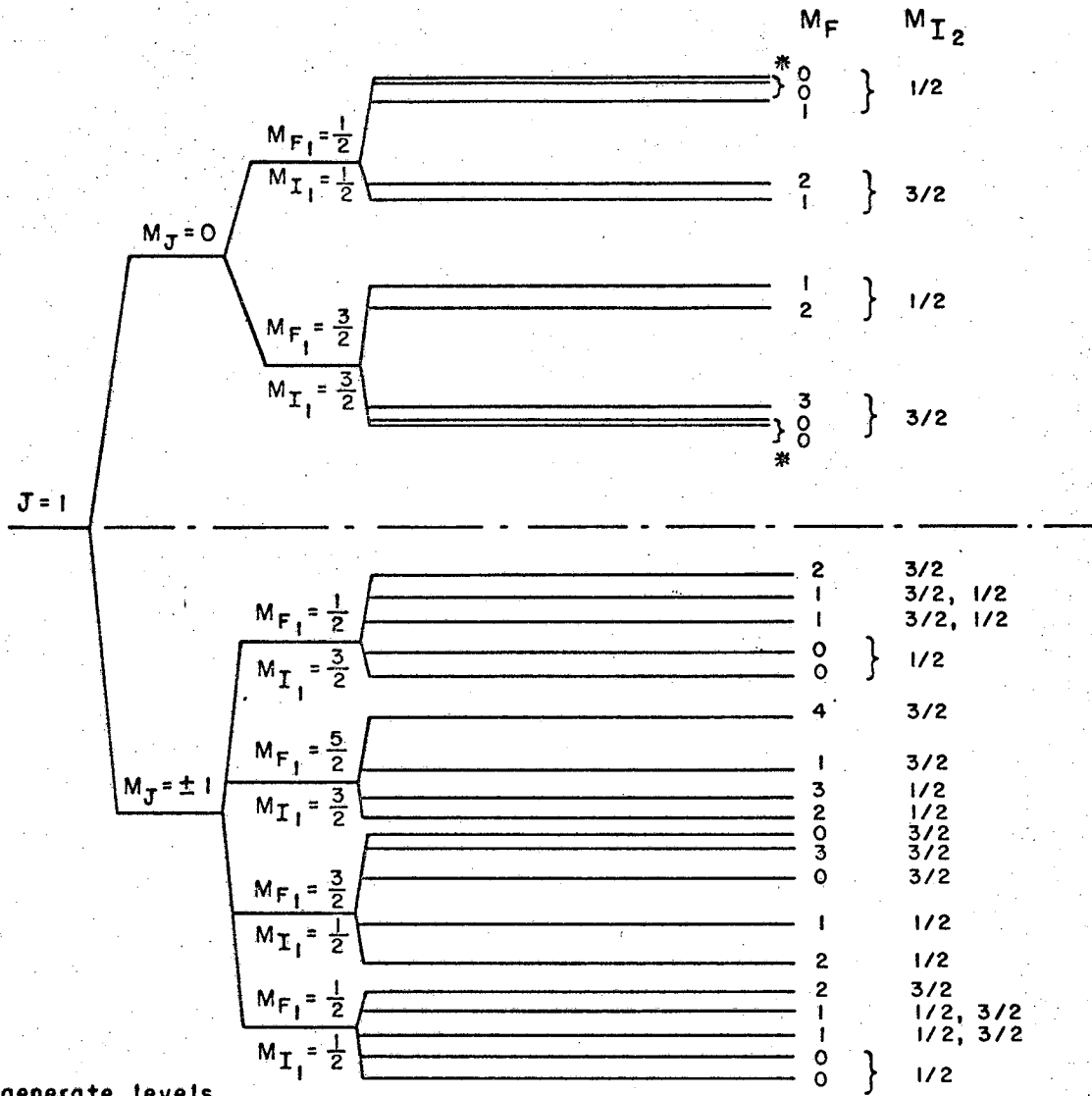
A frequency vs. channel number correlation is then made using the program coincidence scaler in conjunction with the reference voltage generator and oscilloscope. This gives the frequency corresponding to any desired channel of the multi-channel analyzer.

By operating the system for about 1.5 to 2.5 hours a signal to noise ratio of about 5:1 was obtained for most of the lines measured.

### C. Radio-frequency Spectra of NaCl and NaBr.

The spectral lines observed were produced by transitions of the type  $(J = 1, M_J = \pm 1) \rightarrow (J = 1, M_J = 0)$ . The  $J = 1$  level is three-fold degenerate in the absence of external fields and nuclear hyperfine interactions. However, due to the spin of  $3/2$  in both the Na and Cl nuclei, the rotational angular momentum of the molecule is coupled to the spin angular momenta of the nuclei through the nuclear electric quadrupole and spin-rotation interactions. The application of an electric field causes further splitting of the energy levels due to the Stark effect. At a field of 600 v/cm, in which the spectra were taken, the Stark effect is the dominant interaction (Strong field)<sup>16</sup> and the hyperfine interactions can be treated as perturbations on the Stark levels. An energy level diagram for Na<sup>23</sup>Cl<sup>35</sup> at strong fields is shown in Fig. 3.

The hyperfine components of the  $(J = 1, M_J = \pm 1) \rightarrow (J = 1, M_J = 0)$  transition were also investigated in the NaBr molecules at a field strength



\* Degenerate levels  
 a - Schematic; energy level separations not drawn to scale.

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Fig. 3. Energy level diagram<sup>a</sup> for Na<sup>23</sup>Cl<sup>35</sup> at strong fields.

of 600 v/cm. The two Br isotopes have spins equal also to  $3/2$ . A radio-frequency voltage of about 1 volt rms was used to induce transitions between the various levels of the molecules.

### III. RESULTS AND DISCUSSION

The spectra of the molecules were interpreted using the Hamiltonian:<sup>17,18</sup>

$$\begin{aligned}
 H = & B\underline{J}^2 - \underline{\mu} \cdot \underline{E} - eq_1 Q_1 \frac{[3(\underline{I}_1 \cdot \underline{J})^2 + 3/2 (\underline{I}_1 \cdot \underline{J}) - (\underline{I}_1^2 \underline{J}^2)]}{2I_1(2I_1-1)(2J-1)(2J+3)} \\
 & - eq_2 Q_2 \frac{[3(\underline{I}_2 \cdot \underline{J})^2 + 3/2 (\underline{I}_2 \cdot \underline{J}) - (\underline{I}_2^2 \underline{J}^2)]}{2I_2(2I_2-1)(2J-1)(2J+3)} + c_1 (\underline{I}_1 \cdot \underline{J}) + c_2 (\underline{I}_2 \cdot \underline{J}) \\
 & + c_3 \frac{[3(\underline{I}_1 \cdot \underline{J})(\underline{I}_2 \cdot \underline{J}) + 3(\underline{I}_2 \cdot \underline{J})(\underline{I}_1 \cdot \underline{J}) - 2(\underline{I}_1 \cdot \underline{I}_2) J(J+1)]}{(2J-1)(2J+3)} + c_4 (\underline{I}_1 \cdot \underline{I}_2)
 \end{aligned}$$

The first term in the above expression represents the rotational energy of the molecule, where  $B$  is the rotational constant and  $\underline{J}$  the rotational angular momentum operator. The second term represents the interaction of the permanent electric dipole moment  $\underline{\mu}$  with the externally applied electric field  $\underline{E}$ . The third and fourth terms represent the interaction of the nuclear quadrupole moments ( $Q_1$  and  $Q_2$ ) with the field gradients at the nuclei ( $q_1$  and  $q_2$ ).  $I_1$  and  $I_2$  are the nuclear spins,  $e$  is the electronic charge and  $eq_1 Q_1$  and  $eq_2 Q_2$  are the quadrupole coupling constants. The fifth and sixth terms represent the magnetic coupling of the nuclear spins with the molecular angular momentum, where  $c_1$  and  $c_2$  are the spin-rotation interaction constants. The seventh and eighth terms represent the tensor and scalar spin-spin interactions respectively, with  $c_3$  and  $c_4$  the respective interaction constants.<sup>19</sup>

A computer program was used to calculate the matrix elements of the above Hamiltonian using a  $(J, I_1, I_2, M_J, M_{I_1}, M_{I_2})$  representation, where  $M_J$ ,  $M_{I_1}$  and  $M_{I_2}$  are the projections of  $J$ ,  $I_1$  and  $I_2$  respectively in the direction of the field  $E$ . The program calculates the energy eigenvalues by diagonalizing this matrix and then calculates the spectral line positions and intensities, corresponding to the given set of input parameters  $B$ ,  $J$ ,  $I_1$ ,  $I_2$ ,  $eq_1 Q_1$ ,  $eq_2 Q_2$ ,  $c_1$ ,  $c_2$ ,  $c_3$ ,  $c_4$  and  $\mu$ , according to the selection rules  $\Delta M_F = 0, \pm 1, \pm 2, \dots$  where  $M_F$  is the projection of the total angular momentum in the direction of the field. The line intensities are calculated from the matrix elements of the dipole moment operator using the calculated wave functions. The unknown interaction constants are varied to obtain the best fit to the observed spectra.

#### A. Spectra of Na<sup>23</sup>Cl<sup>35</sup> and Na<sup>23</sup>Cl<sup>37</sup>

The spectra of Na<sup>23</sup>Cl<sup>35</sup> at 600 v/cm consist of 13 groups of lines for each vibrational state. All of these were observed for the  $v = 0, 1$ , and 2 vibrational states and a few for the  $v = 3$  state. Each group is a composite of several overlapping lines giving a full width at half maximum (FWHM) of about 30 to 60 KHz. Some separation of the components of these lines was evident as the vibrational state increased. The compound lines appear somewhat broader than that calculated based on the average transit time of the molecules in the C-field region and taking into account C-field inhomogeneities. This is probably due to power broadening of the lines caused by using a radio-frequency voltage of about 1 volt rms. The spectra for the  $v = 0$  state of Na<sup>23</sup>Cl<sup>35</sup> is shown in Fig. 4. The observed line positions are shown in



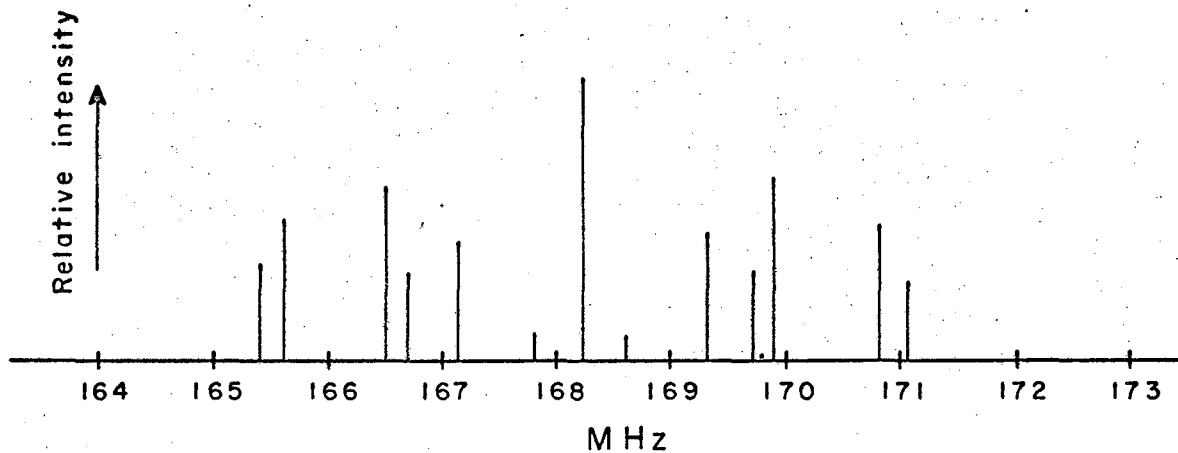


Fig. 4.  $V = 0$  spectra of  $\text{Na}^{23}\text{Cl}^{35}$ .

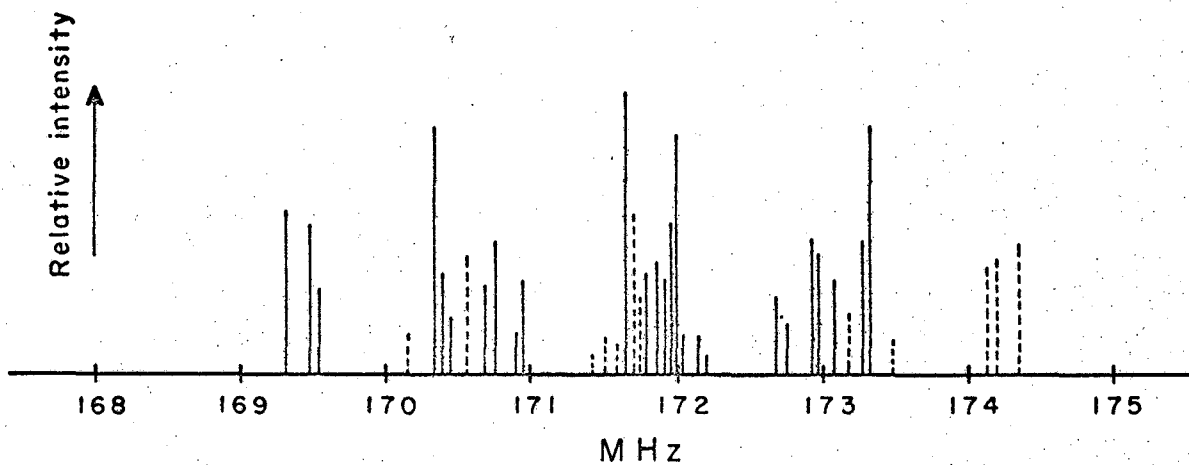


Fig. 5.  $V = 0$  spectra of  $\text{Na}^{23}\text{Cl}^{37}$ .

N.B.-Dotted lines have been overlapped by  $\text{Na}^{23}\text{Cl}^{35}$  lines or have not been observed inadvertently.

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Tables A-II, A-III, A-IV and A-V of the Appendix. The predicted or calculated line frequencies are also given. Most of the lines have been measured individually. The observed line positions for the  $v = 0$  state represent an average of two or more measurements. The NaCl beam was very stable for long periods of time. Fluctuations in beam intensity and oven bursts, as experienced by Gold, have been eliminated by using as oven material sticks of NaCl obtained from previous runs.

In fitting the observed unresolved multiplets, calculated composite line positions were obtained by graphically adding component lines of Gaussian line shapes, with FWHM = 25 KHz and having the calculated frequencies and relative intensities. The centers of these calculated compound lines were then compared with the observed lines until a best fit was obtained. Values of rotational constants used in fitting the spectra were derived from the data of Clouser and Gordy.<sup>29</sup> A sufficiently good fit was obtained using only the dipole moments and the (eqQ)s of the two nuclei. The accuracy of the line measurements did not warrant the use of the small magnetic interaction constants. Magnetic resonance experiments indicate  $c_{Na^{23}}$  to be approximately 1 KHz.<sup>6</sup>  $c_{Cl^{35}}$  is probably of the same order of magnitude as that of  $Cl^{35}$  in TlCl i.e. about 1.4 KHz.<sup>6</sup> This would cause shifts in the spectral lines of 1 to 1.5 KHz at 600 v/cm, which are indeed small, compared to the observed spectral frequencies which ranged from 165 to 171 MHz for the  $v = 0$  state.

Values of the dipole moments and quadrupole coupling constants obtained for a best fit are given in Tables I and II respectively. The dipole moments obtained in the present work are an order of magnitude more accurate than those of Gold. They agree with those of the latter within his error limits. The variation of  $\mu$  with vibrational state for  $Na^{23}Cl^{35}$  has been fitted with a

Table I. Dipole Moments of Na<sup>23</sup>Cl<sup>35</sup> and Na<sup>23</sup>Cl<sup>37</sup>Na<sup>23</sup>Cl<sup>35</sup>

	<u>Present Work</u>	<u>Gold<sup>4</sup></u>
$\mu_0 =$	9.00195 $\pm$ 0.00045 D	9.000 $\pm$ 0.004 D
$\mu_1 =$	9.06190 $\pm$ 0.00046 D	9.061 $\pm$ 0.004 D
$\mu_2 =$	9.12226 $\pm$ 0.00050 D	9.122 $\pm$ 0.004 D
$\mu_3 =$	9.18290 $\pm$ 0.00097 D	
	$\mu_e =$ 8.97208 $\pm$ 0.00065 D	
	$\mu_I =$ 0.05963 $\pm$ 0.00020 D	
	$\mu_{II} =$ 0.00017 $\pm$ 0.00010 D	

Na<sup>23</sup>Cl<sup>37</sup>

$\mu_0 =$	9.0017 $\pm$ 0.00069 D
$\mu_1 =$	9.0610 $\pm$ 0.00071 D
	$\mu_e =$ 8.97205 $\pm$ 0.00130 D
	$\mu_I =$ 0.05930 $\pm$ 0.00060 D

Table II. Quadrupole Coupling Constants in Na<sup>23</sup>Cl<sup>35</sup>

<u>Na<sup>23</sup></u>	<u>Present Work</u>	<u>Gold<sup>4</sup></u>	<u>Cachs et al.<sup>8</sup></u>	<u>Nierenberg<sup>5</sup> and Ramsey</u>	<u>Zeiger<sup>6</sup> and Bolef</u>	<u>Cote<sup>7</sup> and Kusch</u>
$(eqQ)_0$	$= - 5.720 \pm 0.020$ MHz	$- 5.6 \pm 0.1$ MHz	$-5.328 \pm 0.08^*$ MHz	$-5.400^*$ MHz	$-5.520 \pm 0.02^*$ MHz	$-5.670^*$ MHz
$(eqQ)_1$	$= - 5.590 \pm 0.020$ MHz					
$(eqQ)_2$	$= - 5.470 \pm 0.025$ MHz					
$(eqQ)_3$	$= - 5.350 \pm 0.030$ MHz					
$(eqQ)_e$	$= - 5.785 \pm 0.025$ MHz				$-5.608 \pm 0.02$ MHz	
$(eqQ)_{I}$	$= 0.133 \pm 0.005$ MHz					
$(eqQ)_{II}$	$= - 0.003 \pm 0.001$ MHz					
<u>Cl<sup>35</sup></u>						
$(eqQ)_0$	$= - 5.575 \pm 0.020$ MHz					
$(eqQ)_1$	$= - 5.820 \pm 0.020$ MHz					
$(eqQ)_2$	$= - 6.060 \pm 0.025$ MHz					
$(eqQ)_3$	$= - 6.300 \pm 0.030$ MHz					
$(eqQ)_e$	$= - 5.452 \pm 0.025$ MHz					
$(eqQ)_{I}$	$= - 0.247 \pm 0.003$ MHz					
$(eqQ)_{II}$	$= 0.001 \pm 0.001$ MHz					

Values obtained by Molecular Beam Magnetic Resonance method; except for those of Zeiger and Bolef, these values are averaged over the vibrational states of the molecule.

Table III. Quadrupole Coupling Constants in Na<sup>23</sup>Cl<sup>37</sup>Na<sup>23</sup>

$$(\text{eqQ})_0 = - 5.720 \pm 0.010 \text{ MHz}$$

$$(\text{eqQ})_1 = - 5.590 \pm 0.010 \text{ MHz}$$

$$(\text{eqQ})_e = - 5.785 \pm 0.016 \text{ MHz}$$

$$(\text{eqQ})_I = 0.130 \pm 0.015 \text{ MHz}$$

Cl<sup>37</sup>

$$(\text{eqQ})_0 = - 4.370 \pm 0.010 \text{ MHz}$$

$$(\text{eqQ})_1 = - 4.565 \pm 0.010 \text{ MHz}$$

$$(\text{eqQ})_e = - 4.273 \pm 0.016 \text{ MHz}$$

$$(\text{eqQ})_I = - 0.195 \pm 0.015 \text{ MHz}$$

quadratic equation in the vibrational quantum number<sup>11</sup> i.e.

$$\mu_v = \mu_e + \mu_I(v + 1/2) + \mu_{II}(v + 1/2)^2$$

The coefficients  $\mu_e$ ,  $\mu_I$  and  $\mu_{II}$  were obtained by the method of least squares.

Observation of the spectra of  $\text{Na}^{23}\text{Cl}^{37}$  permitted the assignment of the coupling constants in  $\text{Na}^{23}\text{Cl}^{35}$  to the respective nuclei. Since the values  $(\text{eqQ})$  in  $\text{Na}^{23}\text{Cl}^{35}$  are significantly different, that which gives a ratio of  $(\text{eqQ})_{\text{Cl}^{35}}$  to  $(\text{eqQ})_{\text{Cl}^{37}}$  closest to the value of 1.268, observed in many chlorine substituted molecules,<sup>20</sup> corresponds to the  $\text{Cl}^{35}$  coupling.

Results of magnetic resonance measurements, themselves at variance with each other, are somewhat lower than  $(\text{eqQ})_{\text{Na}^{23}}$  observed in this work. Oachs' value for  $(\text{eqQ})_{\text{Cl}^{35}}$  is in agreement with the present work. Gold indicated that his  $V = 0$  spectra is best fitted with coupling constants of 5.5 and 5.7 MHz, with no assignment to the respective nucleus. Within accuracy of his data, however, he concluded that a value of  $5.6 \pm 0.1$  MHz was more likely after making an examination of the calculated values of field gradients in  $\text{Na}^{23}$  and  $\text{Cl}^{35}$  in various molecules.

Spectra of only the  $v = 0$  and 1 vibrational states of  $\text{Na}^{23}\text{Cl}^{37}$  were obtained. Most of the lines were overlapped by the  $\text{Na}^{23}\text{Cl}^{35}$  lines. The lines observed were mostly singlets, the  $(\text{eqQ})$  of the two nuclei being now sufficiently different to cause large separations between the components of each unresolved multiplet line in  $\text{Na}^{23}\text{Cl}^{35}$ . The intensities compared with those of the  $\text{Na}^{23}\text{Cl}^{35}$  lines correspond to that expected for the 25% natural abundance of  $\text{Cl}^{37}$ . The observed spectrum for the  $v = 0$  state of  $\text{Na}^{23}\text{Cl}^{37}$  is shown schematically in Fig. 5.

Dipole moments derived from the spectra are shown in Table I. These are somewhat smaller than those of  $\text{Na}^{23}\text{Cl}^{35}$ . These were fitted with the equation  $\mu = \mu_e + \mu_I(v + 1/2)$ . The equilibrium dipole moments are in excellent agreement. The coefficients  $\mu_I$  in the dipole moment function for both molecules follow the isotopic dependence as predicted by theory,<sup>19</sup> being proportional to  $B_e/\omega_e$ .

The coupling constants for  $\text{Na}^{23}$  in  $\text{Na}^{23}\text{Cl}^{37}$  are essentially the same as those in  $\text{Na}^{23}\text{Cl}^{35}$ . This attests to the correctness of the (eqQ) assignment discussed already. After correcting the chlorine coupling in both NaCl molecules to a common vibrational state the ratio obtained is:

$$\frac{(\text{eq}Q_e)_{\text{Cl}^{35}}}{(\text{eq}Q_e)_{\text{Cl}^{37}}} = 1.2761 \pm 0.0075$$

This value is in good agreement with the observed coupling constants in molecules containing chlorine.<sup>22,23,24,25</sup> It is, however, somewhat higher than more recent microwave measurements<sup>26</sup> on  $\text{CH}_3\text{Cl}$ ,  $\text{ClCN}$ ,  $\text{GeH}_3\text{Cl}$  and the measurements by Wang et al.<sup>27</sup> These give an average value of 1.268. A much closer comparison, however, cannot be made in view of our large error limits brought about by the fact that our spectra had been obtained at strong fields where the line positions are rather insensitive to small changes in the values of the quadrupole interactions.

A weak field experiment would permit a much more accurate determination of these quantities. This will probably be undertaken in this laboratory in the near future.

B. Spectra of Na<sup>23</sup>Br<sup>79</sup> and Na<sup>23</sup>Br<sup>81</sup>

The radio-frequency spectra for the  $J = 1$  and different vibrational states were obtained for Na<sup>23</sup>Br<sup>79</sup> and Na<sup>23</sup>Br<sup>81</sup>. The spectra for each vibrational state consisted of 12 groups of lines as shown in Figs. 6 and 7. Each group is again a composite of a number of lines giving total widths at half maximum of 60 to 80 KHz. Measurements of the line positions are given in Tables A-VIII to A-XIV of the Appendix. Fitting of these spectra was done in the same manner as that of NaCl. Rotational constants used were derived from the data of Rusk and Gordy.<sup>28</sup> Values of dipole moments obtained from an analysis of the spectra are given in Table IV. Quadrupole coupling constants are given in Tables V and VI for Na<sup>23</sup>Br<sup>79</sup> and Na<sup>23</sup>Br<sup>81</sup> respectively. No essential differences could be observed between the dipole moments of the two isotopic molecules. The low signal to noise level of about 2:1 to 3:1 after two hours run time limited the accuracy of the line frequency measurements. The variation of dipole moment with vibrational state is linear within experimental errors. No values for the dipole moments of these molecules have been reported before and no comparison could be made here. The values are, however, what may be expected on the basis of the dipole moments of the other Na-halides.  $(eqQ)_{Na^{23}}$  are the same in both NaBr<sup>79</sup> and NaBr<sup>81</sup>. Our values are fairly close to those obtained by Magnetic resonance (MBMR) methods<sup>7,10</sup> as shown in Table V. The values found for  $(eqQ)_{Br^{79}}$  are in excellent agreement with those obtained by microwave spectroscopy.<sup>9</sup> We have obtained more accurate values, however, and determined the variation of  $(eqQ)$  with vibrational state. A linear variation is obtained. This is also true for  $(eqQ)_{Br^{81}}$  in Na<sup>23</sup>Br<sup>81</sup>. From the equilibrium values of this quantity we obtain:



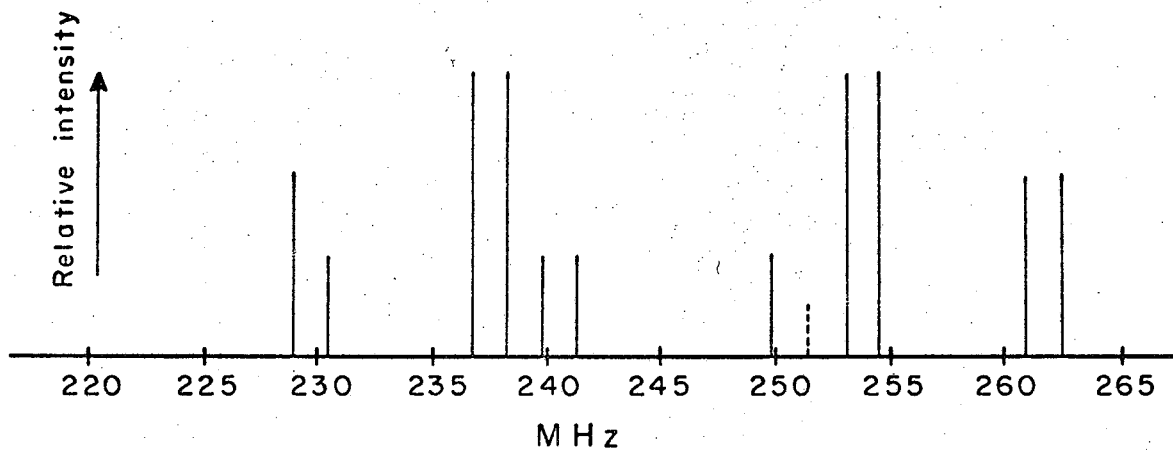


Fig. 6.  $V = 0$  spectra of  $\text{Na}^{23}\text{Br}^{79}$ .

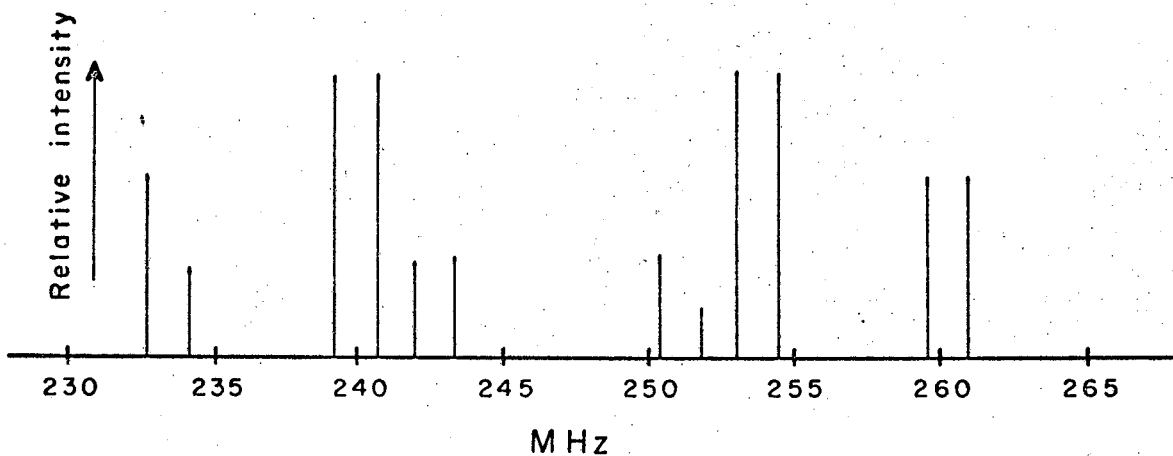


Fig. 7.  $V = 0$  spectra of  $\text{Na}^{23}\text{Br}^{81}$ .

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Table IV. Dipole Moments of Na<sup>23</sup>Br<sup>79</sup> and Na<sup>23</sup>Br<sup>81</sup>Na<sup>23</sup>Br<sup>79</sup>

$$\mu_0 = 9.1183 \pm 0.00056 \text{ D}$$

$$\mu_1 = 9.1715 \pm 0.00062 \text{ D}$$

$$\mu_2 = 9.2246 \pm 0.00063 \text{ D}$$

$$\mu_3 = 9.2779 \pm 0.0007 \text{ D}$$

$$\mu_e = 9.09177 \pm 0.00127 \text{ D}$$

$$\mu_I = 0.05308 \pm 0.00070 \text{ D}$$

$$\mu_{II} = -0.0001 \pm 0.0004 \text{ D}$$

Na<sup>23</sup>Br<sup>81</sup>

$$\mu_0 = 9.1183 \pm 0.00059 \text{ D}$$

$$\mu_1 = 9.1715 \pm 0.00060 \text{ D}$$

$$\mu_2 = 9.2245 \pm 0.00082 \text{ D}$$

$$\mu_e = 9.01978 \text{ D}$$

$$\mu_I = 0.05310 \text{ D}$$

$$\mu_{II} = -0.0001 \text{ D}$$

Table V. Quadrupole Coupling Constants in Na<sup>23</sup>Br<sup>79</sup>

<u>Present Work</u>	<u>Others</u>
<u>Na<sup>23</sup> in Na<sup>23</sup>Br<sup>79</sup></u>	
(eqQ) <sub>0</sub> = -4.820 ± 0.078 MHz	-4.880 MHz (Mol. Beam Mag. Resonance) <sup>7</sup>
	-4.680 MHz ( " " " " ) <sup>10</sup>
(eqQ) <sub>1</sub> = -4.770 ± 0.078 MHz	
(eqQ) <sub>2</sub> = -4.720 ± 0.090 MHz	
(eqQ) <sub>3</sub> = -4.670 ± 0.100 MHz	
(eqQ) <sub>e</sub> = -4.845 MHz	
(eqQ) <sub>I</sub> = 0.050 MHz	
(eqQ) <sub>II</sub> = 0.0 MHz	
<u>Br<sup>79</sup> in Na<sup>23</sup>Br<sup>79</sup></u>	
(eqQ) <sub>0</sub> = 58.060 ± 0.080 MHz	58.000 ± 2.000 MHz (Microwave) <sup>9</sup>
(eqQ) <sub>1</sub> = 59.160 ± 0.110 MHz	
(eqQ) <sub>2</sub> = 60.300 ± 0.100 MHz	
(eqQ) <sub>3</sub> = 61.400 ± 0.150 MHz	
(eqQ) <sub>e</sub> = 57.498 ± 0.130 MHz	
(eqQ) <sub>I</sub> = 1.116 ± 0.037 MHz	
(eqQ) <sub>II</sub> = 0.0 ± 0.009 MHz	

Table VI. Quadrupole Coupling Constants in Na<sup>23</sup>Br<sup>81</sup>Na<sup>23</sup> in Na<sup>23</sup>Br<sup>81</sup>

$(eqQ)_0$	=	- 4.820	±	0.065	MHz
$(eqQ)_1$	=	- 4.770	±	0.075	MHz
$(eqQ)_2$	=	- 4.720	±	0.080	MHz
$(eqQ)_e$	=	- 4.845			MHz
$(eqQ)_I$	=	0.050			MHz
$(eqQ)_{II}$	=	0.0			MHz

Br<sup>81</sup> in Na<sup>23</sup>Br<sup>81</sup>

$(eqQ)_0$	=	48.550	±	0.090	MHz
$(eqQ)_1$	=	49.700	±	0.090	MHz
$(eqQ)_2$	=	50.850	±	0.110	MHz
$(eqQ)_e$	=	47.975			MHz
$(eqQ)_I$	=	1.150			MHz
$(eqQ)_{II}$	=	0.0			MHz

$$\frac{(\text{eqQ})_{\text{Br}^{79}}}{(\text{eqQ})_{\text{Br}^{81}}} = 1.1985 \pm 0.0039$$

This is in agreement, within our experimental error, with values of the ratio observed previously.<sup>30</sup>

The accuracy of the line position measurements did not warrant including the magnetic interactions in fitting the spectra. Line shifts due to these are expected to be insignificant.

APPENDIX

Table A-I.	Electronic Equipment for Signal Averaging System.
Table A-II.	V = 0 Spectra of Na <sup>23</sup> Cl <sup>35</sup>
Table A-III.	V = 1 Spectra of Na <sup>23</sup> Cl <sup>35</sup>
Table A-IV.	V = 2 Spectra of Na <sup>23</sup> Cl <sup>35</sup>
Table A-V.	V = 3 Spectra of Na <sup>23</sup> Cl <sup>35</sup>
Table A-VI.	V = 0 Spectra of Na <sup>23</sup> Cl <sup>37</sup>
Table A-VII.	V = 1 Spectra of Na <sup>23</sup> Cl <sup>37</sup>
Table A-VIII.	Na <sup>23</sup> Br <sup>79</sup> Spectra V = 0
Table A-IX.	Na <sup>23</sup> Br <sup>79</sup> Spectra V = 1
Table A-X.	Na <sup>23</sup> Br <sup>79</sup> Spectra V = 2
Table A-XI.	Na <sup>23</sup> Br <sup>79</sup> Spectra V = 3
Table A-XII.	Na <sup>23</sup> Br <sup>81</sup> Spectra V = 0
Table A-XIII.	Na <sup>23</sup> Br <sup>81</sup> Spectra V = 1
Table A-XIV.	Na <sup>23</sup> Br <sup>81</sup> Spectra V = 2

Table A-I. Electronic Equipment for Signal Averaging System.

<u>Unit</u>	<u>Model No.</u>	<u>Manufacturer</u>
Transistorized linear pulse amplifier		LRL <sup>a</sup> Plan No. 15X4845
Inverting amplifier (gain 1-10X, zero offset)		LRL <sup>a</sup> Plan No. 13X1450
Frequency or pulse scaler (0 to 10 <sup>8</sup> counts per second; with 5253A and 5253B plug-ins the counting range is extended to 500 MHz.)	5245L	Hewlett-Packard Co., Palo Alto, Calif.
Radio frequency signal generators:		Hewlett-Packard Co., Palo Alto, Calif.
a) 50 kHz to 65 MHz.	606B	
b) 10 MHz to 455 MHz.	608F	
Digital recorder with analog output	562A	Hewlett-Packard Co., Palo Alto, Calif.
Frequency synchronizer (locks 606B and 608F signal generators and allows modulation)	8708A	Hewlett-Packard Co., Palo Alto, Calif.,
Frequency mixer (50 kHz to 450 MHz.)	10514A	Hewlett-Packard Co.,
Stark field voltage supply (1-6000V at 20 mA)	HV-1556	Power Designs Pacific, Inc., Palo Alto, Calif.
Reference voltage supply (0-10 volts)		LRL <sup>a</sup> Plan No. 13X1470
Ramp voltage generator		LRL <sup>a</sup> Plan No. 13X1430
Start pulse function generator	301	Electronics, Inc. Portland, Oregon
Single channel analyzer	NC-11	Hammer Electronics Co. Inc., Princeton, New Jersey
Multichannel analyzer (1024 channels in quadrants, 2x10 <sup>-5</sup> sec dead time per channel)	NS-610	Northern Scientific Madison, Wisconsin
Data processor	460	Northern Scientific Madison, Wisconsin
Switch programed time based oscillator		LRL <sup>a</sup> Plan No. 15X8410
Programed coincidence scaler (0-9999 channels)		LRL <sup>a</sup> Plan No. 13X1310

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Table A-II.  $V = 0$  Spectra of  $\text{Na}^{23}\text{Cl}^{35}$   
 $E = 600 \text{ v/cm}$

Observed Line <sup>a</sup> Positions	Calculated Line Positions			Difference
	Calc'd Freq.	Intensity	Composite Line Frequency	
1. 165,433.7 KHz	165,433.3 KHz	59.85	165,433.5 KHz	-0.2 KHz
2. 165,634.2	165,628.7	23.84	165,632.0	-2.2
	165,628.7	24.09		
	165,635.4	23.37		
	165,635.4	23.41		
3. 166,548.8	166,531.7	19.71	166,550.0 <sup>b</sup>	1.2
	166,546.5	20.41		
	166,559.5	100.00		
4. 166,736.5	166,725.2	4.25	166,737.5	1.0
	166,731.9	38.89		
	166,743.8	38.40		
	166,750.4	4.25		
5. 167,115.2	167,089.9	15.53	167,116.0 <sup>b</sup>	0.8
	167,096.6	15.49		
	167,104.7	14.44		
	167,110.7	26.48		
	167,110.7	17.37		
	167,111.5	14.39		
	167,137.4	16.65		
	167,137.4	25.33		
6. 167,835.9	167,840.3	4.71	167,843.5	7.6
	167,840.3	4.67		
	167,846.9	5.07		
	167,846.9	4.98		
7. 168,230.6	168,203.2	20.01	168,230.0	-0.6
	168,207.2	13.51		
	168,209.8	20.10		
	168,213.6	98.64		
	168,219.5	51.43		
	168,219.8	24.27		
	168,227.6	1.35		
	168,230.9	24.27		
	168,232.1	1.34		
	168,234.3	48.55		
	168,234.6	25.72		
	168,245.8	25.71		
	168,246.1	98.63		
	168,252.5	13.51		
	168,253.2	40.13		



Table A-II. (cont'd)

Observed Line <sup>a</sup> Positions	Calculated Line Positions			Difference
	Calc'd Freq.	Intensity	Composite Line Frequency	
8. 168,616.6 KHz	168,612.7 KHz	4.98	168,616.0 KHz	-0.6
	168,612.7	5.07		
	168,619.4	4.67		
	168,619.4	4.72		
9. 169,346.7	169,322.3	25.32	169,343.5 <sup>b</sup>	-3.2
	169,322.3	16.64		
	169,349.0	17.36		
	169,349.0	26.47		
	169,351.6	28.84		
	169,366.4	31.02		
10. 169,723.5	169,709.3	4.26	169,721.5	-2.0
	169,715.9	38.41		
	169,727.8	38.89		
	169,734.4	4.25		
11. 169,916.6	169,900.2	99.95	169,905.5 <sup>b</sup>	-7.1
	169,911.0	10.19		
	169,915.4	10.23		
	169,925.8	9.83		
	169,930.2	9.88		
12. 170,826.4	170,824.3	23.40	170,827.5	1.1
	170,824.3	23.37		
	170,830.9	24.09		
	170,830.9	23.84		
13. 171,025.5	171,024.2	29.97	171,026.0	0.5
	171,028.6	29.88		

<sup>a</sup> Average error in observed line positions = 2.4 KHz

<sup>b</sup> Center of composite line, does not necessarily coincide with peak.

Table A-III.  $V = 1$  Spectra of  $\text{Na}^{23}\text{Cl}^{35}$   
 $E = 600 \text{ v/cm}$

Observed Line <sup>a</sup> Positions	Calculated Line Positions			Difference
	Calc'd Freq <sub>1</sub>	Intensity	Composite Line Frequency	
1. 168,868.1 KHz	168,870.5 KHz	59.86	168,870.5 KHz	2.4 KHz
2. 169,072.7	169,066.0	24.02	169,071.5	-1.2
	169,066.0	24.24		
	169,076.3	23.17		
	169,076.3	23.25		
3. 169,969.0 170,006.8	169,971.4	19.51	169,971.4	2.4
	170,002.3	20.61	170,007.5	0.7
	170,008.3	100.00		
4. 170,185.2 <sup>b</sup>	170,165.9	1.25	170,187.5	2.3
	170,176.2	42.06		
	170,198.8	41.26		
	170,209.1	1.25		
5. 170,563.5 <sup>b</sup>	170,534.9	15.85	170,570.0 <sup>c</sup>	6.5
	170,541.7	25.11		
	170,557.3	19.27		
	170,565.9	14.12		
	170,572.6	14.06		
	170,599.6	18.03		
6. 171,299.1	171,298.7	4.62	171,304.5	5.4
	171,298.7	4.57		
	171,309.0	5.16		
	171,309.0	5.08		
7. 171,668.6  171,693.2(ctr)  171,724.2 <sup>b</sup>	171,657.2	13.99	171,671.5  171,695.0(ctr)  171,718.5	2.9  1.8(ctr)  -5.7
	171,666.8	20.01		
	171,667.4	99.79		
	171,673.5	20.10		
	171,676.1	52.28		
	171,677.0	23.84		
171,688.1	23.85			
171,724.2 <sup>b</sup>	171,707.0	47.70	171,718.5	-5.7
	171,707.9	26.15		
	171,719.0	26.13		
	171,719.5	40.13		
	171,722.3	99.78		
	171,732.3	14.00		

Table A-III. (cont'd)

Observed Line <sup>a</sup> Positions	Calculated Line Positions			Difference	
	Calc'd Freq.	Intensity	Composite Line Frequency		
8. 172,090.6 KHz	172,080.5 KHz	5.09	172,085.5	-5.1	
	172,080.5	5.17			
	172,090.8	4.58			
	172,090.8	4.63			
9. 172,818.5	172,790.0	23.43	172,819.5 <sup>c</sup>	1.0	
	172,790.0	18.01			
	172,820.4	28.19			
	172,832.3	19.27			
	172,832.3	25.10			
	172,851.3	31.67			
10. 173,209.3 <sup>b</sup>	173,180.5	1.25	173,202.5	-6.8	
	173,190.8	41.27			
	173,213.3	42.06			
	173,223.6	1.25			
11. 173,383.4 <sup>d</sup>	173,381.3	99.95	173,382.5	-0.9	
	173,385.1	10.29			
	173,389.5	10.33			
		173,416.0	9.73	173,416.5 <sup>b</sup>	
		173,420.4	9.78		
12. 174,314.6	174,313.3	23.24	174,318.5	3.9	
	174,313.3	23.18			
	174,323.5	24.23			
	174,323.5	24.02			
13. 174,513.8	174,516.9	29.97	174,519.0	5.2	
	174,521.3	29.88			

<sup>a</sup>Average line error = 3.2 KHz

<sup>b</sup>Overlaps with  $V = 0$  Na<sup>23</sup>Cl<sup>37</sup> lines.

<sup>c</sup>Center of composite line(s) does not necessarily coincide with peak(s).

<sup>d</sup>Fitted main peak; smaller peak overlaps with  $V = 0$  Na<sup>23</sup>Cl<sup>37</sup> line.

Table A-IV. V = 2 Spectra of Na<sup>23</sup>Cl<sup>35</sup>  
E = 600 v/cm

Observed Line Positions	Calculated Line Positions		Composite Line <sup>a</sup> Frequency	Difference
	Calc'd Freq.	Intensity		
1. 172,376.1 KHz	173,377.5 KHz	59.87	172,377.5 KHz	1.4 KHz
2. 172,584.5	172,576.1	24.72	172,579.5	-5.0
	172,567.1	24.88		
	172,592.7	22.36		
	172,592.7	22.49		
3. 173,539.2 <sup>b</sup>	173,454.3	18.67	173,538.5 <sup>2</sup>	-0.7
	173,527.9	100.00		
	173,555.0	21.42		
4. 173,670.4	173,669.2	43.87	173,669.2	-1.2
173,739.3	173,744.9	41.79	173,744.9	5.6
5. 174,048.5	174,021.2	17.23	174,037.5	-11.0
	174,027.7	17.21		
	174,049.7	24.55		
	174,049.7	22.16		
174,110.6 <sup>c</sup>	174,121.9	12.77	174,125.5	14.9
	174,128.5	12.69		
174,155.4 <sup>c</sup>	174,158.3	18.64	174,158.3	12.9
	174,158.3	20.60		
6. 174,830.3	174,821.5	4.25	174,835.0	4.7
	174,821.5	4.19		
	174,847.0	5.60		
	174,847.0	5.53		
7. 175,180.1	175,126.2	14.03	175,171.5	-8.6
	175,151.7	99.98		
	175,174.9	55.84		
	175,181.2	22.04		
	175,192.2	22.06		
	175,198.7	19.98		
	175,205.3	20.07		
175,231.1(ctr)			175,231.0(ctr)	-0.1
175,277.7	175,260.4	40.08	175,290.5	12.8
	175,275.7	44.10		
	175,281.9	27.94		
	175,292.9	27.90		
	175,310.7	99.97		
	175,336.2	14.04		

Table A-IV. (cont'd)

Observed Line Positions	Calculated Line Positions		Composite Line <sup>a</sup> Frequency	Difference
	Calc'd Freq.	Intensity		
8. 175,633.6	175,615.3 KHz	5.53	175,627.0	-6.6
	175,615.3	5.61		
	175,640.9	4.19		
	175,640.9	4.25		
9. -- <sup>d</sup>	176,304.1	20.60	176,319.0 <sup>a</sup>	--
	176,304.1	18.63		
	176,337.2	25.47		
176,413.3	176,412.7	22.15	176,424.0 <sup>a</sup>	10.7
	176,412.7	24.54		
	176,437.9	34.45		
10. 176,718.1	176,717.4	41.81	176,717.4	-0.7
176,791.2 <sup>c</sup>	176,793.2	43.88	176,793.2	2.0
11. 176,921.3 <sup>f</sup>	176,905.2	10.69	176,925.0 <sup>a</sup>	3.7
	176,909.5	10.74		
	176,934.4	99.95		
177,011.3	177,005.9	9.31	177,007.5	-3.6
	177,010.3	9.36		
12. 177,877.7	177,869.7	22.48	177,882.5	4.8
	177,869.7	22.36		
	177,895.2	24.88		
	177,895.2	24.72		
13. 178,082.9	178,082.7	29.98	178,084.5	1.6
	178,087.1	29.89		

Average line error = 5.1 KHz

<sup>a</sup> Composite line center does not necessarily coincide with peak(s).

<sup>b</sup> Smaller peak overlapped by  $V = 0$  Na<sup>23</sup>Cl<sup>37</sup> line.

<sup>c</sup> Overlap with  $V = 0$  Na<sup>23</sup>Cl<sup>37</sup> lines.

<sup>d</sup> Overlap with  $V = 3$  Na<sup>23</sup>Cl<sup>35</sup> line.

<sup>e</sup> Overlap with  $V = 1$  Na<sup>23</sup>Cl<sup>35</sup> line.

<sup>f</sup> Overlap with  $V = 1$  Na<sup>23</sup>Cl<sup>37</sup> line.

Table A-V.  $V = 3$  Spectra of  $\text{Na}^{23}\text{Cl}^{35}$   
 $E = 600 \text{ v/cm}$

Observed Line Positions	Calculated Line Positions		Composite Line Frequency <sup>a</sup>	Difference
	Calc'd Freq.	Intensity		
1. 175,959.2 KHz	175,951.8 KHz	59.91	175,951.8 KHz	-7.4 KHz
2. 176,155.7 <sup>b</sup>	176,135.2	25.35	176,155.0	-0.7
	176,135.2	25.49		
	176,175.8	21.54		
	176,175.8	21.68		
3. 177,013.6 177,138.0 177,170.3	177,004.5	17.85	177,004.5	-9.1
	177,115.6	100.00	177,115.6	-22.4
	177,174.9	22.22	177,174.9	4.6
4. 177,251.6 177,359.3	177,228.3	44.68	177,228.3	-23.3
	177,358.6	41.35	177,358.6	-0.7
5. -- <sup>c</sup>				
6. --				
7. --				
8. --				
9. --				
10. 180,315.9 180,441.5	180,312.1	41.36	180,312.1	-3.8
	180,442.4	44.68	180,442.4	0.9
11. 180,493.4 180,556.9 180,664.9	180,493.6	11.10	180,495.5	2.1
	180,497.9	11.13		
	180,555.2	99.95	180,555.2	-1.7
180,664.9	180,664.0	8.89	180,666.0	1.1
	180,668.4	8.95		
12. 181,491.9 --	181,494.9	21.67	181,494.9	3.0
	181,494.9	21.54		
--	181,535.5	25.49	181,535.5	--
	181,535.5	25.34		
13. 181,713.2	181,716.8	29.99	181,719.0	5.8
	181,721.2	29.90		

Average line error = 6.5 KHz

<sup>a</sup>Composite line center does not necessarily coincide with peak(s).

<sup>b</sup>Overlap with  $V = 1$   $\text{NaCl}^{37}$  line.

<sup>c</sup>Lines have not been observed; calculated line positions have thus been omitted.

Table A-VI. V = 0 Spectra of Na<sup>23</sup>Cl<sup>37</sup>  
E = 600 v/cm

Observed Line Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
1. 169,303.0 KHz	169,302.1 KHz	60.01	169,302.1 KHz	-0.9 KHz
2. 169,450.6	169,448.4 169,448.4	26.33 26.45	169,448.4	-2-2
3. 169,501.8	169,504.9 169,504.9	20.03 20.16	169,504.9	3.1
4. -- <sup>b</sup>	170,162.5	16.41	170,162.5	
5. 170,316.9	170,313.3	100.00	170,313.3	-3.6
6. 170,365.4	170,354.3	46.06	170,365.3	-0.1
7. 170,407.0	170,413.9	23.56	170,413.9	6.9
8. --	170,560.4	40.55	170,560.4	
9. 170,649.1	170,643.1 170,647.9	20.74 20.77	170,645.5	-3.6
10. 170,708.2	170,693.7 170,693.7	27.05 26.13	170,693.7	-14.5
11. 170,889.9	170,894.6 170,899.3	9.42 9.33	170,897.0	7.1
12. 170,931.7	170,940.5 170,940.5	16.47 17.01	170,940.5	8.8
13. -- <sup>c</sup>	171,420.8 171,420.8	3.34 3.29	171,420.8	
14. -- <sup>c</sup>	171,477.3 171,477.3	6.89 6.83	171,477.3	
15. -- <sup>c</sup>	171,554.1	13.33	171,554.1	
16. 171,607.7	171,610.6	99.98	171,610.6	2.9
17. -- <sup>b</sup>	171,656.7	64.82	171,656.7	
18. -- <sup>b</sup>	171,694.4 171,702.5	17.43 17.48	171,698.5	
19. 171,762.9	171,754.9 171,759.8	19.82 19.89	171,757.5	-5.4
20. 171,850.8	171,849.2	39.72	171,849.2	-1.6
21. 171,908.9	171,908.2	34.91	171,908.2	-0.7

Table A-VI. (cont'd)

Observed Line Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
22. 171,951.4 KHz	171,945.8 KHz 171,953.9	32.43 32.38	171,950.0 KHz	-1.4 KHz
23. 171,993.5	171,996.1	99.97	171,996.1	2.6
24. 172,042.0	172,052.5	13.33	172,052.5	10.5
25. 172,133.7	172,129.3 172,129.3	6.83 6.70	172,129.3	-4.4
26. 172,194.3	172,185.8 172,185.8	3.29 3.34	172,185.8	-8.5
27. 172,674.3	172,666.0 172,666.0	17.01 16.46	172,666.0	-8.3
28. 172,715.3	172,709.6	18.76	172,709.6	-5.7
29. 172,903.5	172,912.9 172,912.9	26.13 27.03	172,912.9	9.4
30. 172,961.7	172,961.1	41.51	172,961.1	-0.6
31. 173,051.2	173,046.2	40.56	172,046.2	-5.0
32. -- <sup>b</sup>	173,190.9 173,194.3	11.77 11.79	173,192.5	
33. 173,245.9	173,241.3	46.07	173,241.3	-4.6
34. 173,294.1	173,293.3	99.96	173,293.3	-0.8
35. -- <sup>b</sup>	173,442.4 173,445.8	8.18 8.23	173,444.0	
36. -- <sup>d</sup>	174,101.7 174,101.7	20.15 20.02	174,101.7	
37. -- <sup>d</sup>	174,158.1 174,158.2	26.45 26.32	174,158.2	
38. -- <sup>b</sup>	174,302.9 174,306.2	30.04 29.96	174,304.5	

<sup>a</sup>Average line error = 8.8 KHz.

<sup>b</sup>Lines overlap with  $V = 1$  Na<sup>23</sup>Cl<sup>35</sup> lines.

<sup>c</sup>Lines have not been observed, inadvertently.

<sup>d</sup>Overlap with  $V = 2$  Na<sup>23</sup>Cl<sup>35</sup> lines.



Table A-VII. V = 1 Spectra of Na<sup>23</sup>Cl<sup>37</sup>  
E = 600 v/cm

Observed Line Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
1. -- a	172,785.8 KHz	59.95	172,785.8 KHz	
2. -- f	172,941.1	25.73	172,941.1	
	172,941.1	25.86		
3. -- f	172,984.6	20.99	172,984.6	
	172,984.6	21.12		
4. -- b	173,684.3	17.31	172,684.3	
5. 173,795.1	173,801.5	100.00	173,801.5	6.4
6. 173,870.4	173,871.7	22.72	173,879.0	8.6
	173,882.9	45.19		
7. -- b	174,027.0	41.05	174,027.0	
8. -- b	174,175.9	19.38	174,178.5	
	174,180.8	19.39		
9. 174,212.2	174,214.4	25.85	174,214.4	2.2
	174,214.4	24.71		
10. -- c	174,363.2	10.70	174,366.0	
	174,368.2	10.61		
11. 174,407.6	174,402.6	17.49	174,402.6	-5.0
	174,402.6	18.26		
12. -- d	174,925.4	3.68	174,925.4	
	174,925.4	3.63		
13. -- d	174,968.9	6.35	174,968.9	
	174,968.9	6.29		
14. -- b	175,112.8	13.68	175,112.8	
15. -- b	175,156.3	99.98	175,156.3	
16. -- b	175,194.1	61.36	175,194.1	
17. -- b	175,224.0	19.26	175,219.0	
	175,215.7	19.21		
18. -- b	175,261.7	19.90	175,264.0	
	175,266.6	19.97		
19. -- b	175,337.1	39.89	175,337.1	
20. -- b	175,381.4	38.47	175,381.4	

Table A-VII. (cont'd)

Observed Line Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
21. 175,406.1 KHz	175,403.1 KHz 175,411.3	30.69 30.66	175,407.0 KHz	0.9
22. 175,439.0	175,445.0	99.98	175,445.0	6.0
23.	175,488.5	13.69	175,488.5	-8.3
24. -- a	175,632.4 175,632.4	6.29 6.36	175,632.4	
25. -- d	175,675.9 175,675.9	3.63 3.68	175,675.9	
26. 176,192.7	176,198.7 176,198.7	18.26 17.48	176,198.7	6.0
27. 176,235.4	176,235.6	21.31	176,235.6	0.2
28. -- b	176,386.8 176,386.8	24.70 25.84	176,386.8	
29. -- b	176,422.9	38.79	176,422.9	
30. 176,570.2	176,574.2	41.06	176,574.2	4.0
31. -- b	176,718.3 176,727.9 176,731.3	45.19 11.35 11.38	176,721.0	
32. -- b	176,799.7	99.96	176,799.7	
33. -- b	176,915.3 176,918.6	8.63 8.68	176,917.0	
34. -- e	177,616.6 177,616.6	21.12 20.99	177,616.6	
35. -- e	177,660.1 177,660.1	25.86 25.73	177,660.1	
36. 177,812.9	177,813.7 177,817.1	30.01 29.93	177,815.5	2.6

Average line error = 7.3 KHz

<sup>a</sup>Overlap with V = 1 Na<sup>23</sup>Cl<sup>35</sup> line(s).<sup>b</sup>Overlap with V = 2 Na<sup>23</sup>Cl<sup>35</sup> line(s).<sup>c</sup>Overlap with V = 1 Na<sup>23</sup>Cl<sup>35</sup> and V = 0 Na<sup>23</sup>Cl<sup>37</sup> lines.<sup>d</sup>Lines have not been observed.<sup>e</sup>Overlap with V = 3 Na<sup>23</sup>Cl<sup>35</sup> line(s).<sup>f</sup>Overlap with V = 0 Na<sup>23</sup>Cl<sup>37</sup> line(s).

Table A-VIII. Na<sup>23</sup>Br<sup>79</sup> Spectra V = 0  
 (J=1, M<sub>J</sub>=±1, V=0  $\xrightarrow{E=600 \text{ v/cm}}$  J=1, M<sub>J</sub>=0, V=0)

Observed Line <sup>a</sup> Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
1. 228,954 KHz	228,952 KHz 228,988	63.99 64.03	228,970 KHz	16 KHz
2. 230,357	230,352 230,395 230,398 230,398	31.93 31.96 31.98 32.01	230,375	18
3. 236,706	236,694 236,734 236,745	100.00 49.53 49.49	236,720	14
4. 238,121	238,106 238,139 238,139	99.98 49.64 49.54	238,123	2
5. 239,924	239,915 239,915 239,951	17.64 17.62 35.73	239,933	9
6. 241,339	241,315 241,322 241,361	17.56 17.58 35.73	241,338	- 1
7. 249,964	249,937 249,980	36.08 35.49	249,958	- 6
8. -- <sup>b</sup>	251,345 251,349 251,383 251,383	18.03 18.03 17.78 17.81	251,364	
9. 253,156	253,158 253,158 253,191	49.53 49.63 99.97	253,175	19
10. 254,566	254,559 254,604	98.98 99.95	254,581	15
11. 260,934	260,899 260,899 260,941	31.93 31.91 63.73	260,921	-13
12. 262,330	262,308 262,312 262,346	31.93 31.92 63.80	262,327	- 3

<sup>a</sup>Average line error = 19 KHz.

<sup>b</sup>Line has not been observed inadvertently.

Table A-IX. Na<sup>23</sup>Br<sup>79</sup> Spectra V = 1  
 (J=1, M<sub>J</sub>=±1, V=L  $\xrightarrow{E=600 \text{ v/cm}}$  J=1, M<sub>J</sub>=0, V=1)

Observed Line <sup>a</sup> Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
1. 232,931 KHz	232,944 KHz 232,979	63.99 64.04	232,962 KHz	31 KHz
2. 234,312	234,329 234,337 234,373 234,373	31.93 31.97 31.99 32.01	234,351	39
3. 240,841	240,830 240,868 240,879	100.00 49.56 49.52	240,855	14
4. 242,232	242,227 242,259 242,259	99.98 49.66 49.56	242,243	10
5. 244,084	244,099 244,099 244,134	17.65 17.63 35.73	244,117	33
6. 245,485	245,484 245,491 245,528	17.59 17.59 35.73	245,507	22
7. 254,369 <sup>b</sup>	254,321 254,361	36.08 35.53	254,341	-28
8. 255,701	255,714 255,717 255,749 255,749	18.04 18.03 17.79 17.82	255,732	31
9. 257,582	257,589 257,589 257,621	49.55 49.64 99.96	257,606	24
10. 258,981	258,977 259,019	99.03 99.94	258,998	17
11. 265,464	265,476 265,476 265,516	31.93 31.91 63.74	265,496	32
12. -- <sup>c</sup>	266,869 266,872 266,905	31.93 31.92 63.81	266,887	

<sup>a</sup>Average line error = 30 KHz.

<sup>b</sup>Overlap with V = Na<sup>23</sup>Br<sup>81</sup> line.

<sup>c</sup>Line has not been observed inadvertently

Table A-X. Na<sup>23</sup>Br<sup>79</sup> Spectra V = 2  
 (J=1, M<sub>J</sub>=±1, V=2  $\xrightarrow{E=600 \text{ v/cm}}$  J=1, M<sub>J</sub>=0, V=2)

Observed Line <sup>a</sup> Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
1. 236,965 KHz	236,976 KHz 267,009	64.00 64.04	236,993 KHz	28 KHz
2. 238,411	238,346 238,353 238,389 238,389	31.94 31.97 31.99 32.01	238,368	-43
3. 245,014	245,011 245,048 245,058	100.00 49.59 49.55	245,035	21
4. 246,423	246,393 246,424 246,424	99.98 49.68 29.58	246,408	-15
5. -- <sup>b</sup>	248,331 248,331 248,364	17.66 17.64 35.72	248,347	
6. -- <sup>b</sup>	249,701 249,708 249,743	17.60 17.61 35.73	249,722	
7. 258,797 <sup>c</sup>	258,759 258,798	36.08 35.57	258,778	-19
8. 260,140	260,136 260,140 260,171 260,171	18.04 18.04 17.81 17.83	260,154	14
9. 262,086	262,078 262,078 262,109	49.57 49.66 99.96	262,093	7
10. 263,462	263,450 263,491	99.08 99.94	263,471	9
11. -- <sup>b</sup>	270,113 270,113 270,152	31.93 31.91 63.75	270,133	
12. 271,503	271,491 271,494 271,526	31.93 31.92 63.82	271,508	5

<sup>a</sup> Average line error = 26 KHz.

<sup>b</sup> Line has not been observed, inadvertently or sufficiently well.

<sup>c</sup> Overlap with V = 1 Na<sup>23</sup>Br<sup>81</sup> line.

Table A-IX. Na<sup>23</sup>Br<sup>79</sup> Spectra V = 3  
 (J=1, M<sub>J</sub>=±1, V=3  $\xrightarrow{E=600 \text{ v/cm}}$  J=1, M<sub>J</sub>=0, V=3)

Observed Line <sup>a</sup> Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
1. 241,097 KHz	241,086 KHz 241,118	64.01 64.05	241,102 KHz	5 KHz
2. 242,459	242,441 242,448 242,481 242,482	31.95 31.98 31.99 32.02	242,462	3
3. -- b	249,265 249,300 249,310	100.00 49.61 49.58	249,288	
4. -- b	250,631 250,661 250,661	99.98 49.69 49.61	250,646	
5. 252,654	252,631 252,631 252,663	17.67 17.65 35.72	252,647	- 7
6. -- b	253,987 253,993 254,027	17.62 17.62 35.72	254,007	
7. -- b	263,260 263,297	36.09 35.60	263,278	
8. 264,652	264,623 264,626 264,656 264,656	18.04 18.04 17.82 17.86	264,639	-13
9. 266,619	266,626 266,626 266,656	49.59 49.67 99.95	266,641	22
10. 268,010	267,984 268,023	99.12 99.93	268,004	- 6
11. -- b	274,805 274,805 274,842	31.94 31.92 63.77	274,824	
12. -- b	276,168 276,171 276,201	31.94 31.93 63.82	276,185	

<sup>a</sup>Average line error = 39 KHz.

<sup>b</sup>Line has not been observed inadvertently or sufficiently well.

Table A-XIII. Na<sup>23</sup>Br<sup>81</sup> Spectra V = 0  
 (J=1, M<sub>J</sub>=±1, V=0  $\xrightarrow{E=600 \text{ v/cm}}$  J=1, M<sub>J</sub>=0, V=0)

Observed Line <sup>a</sup> Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
1. 232,800 KHz	232,765 KHz 232,807	63.84 63.78	232,786 KHz	-14 KHz
2. 234,187	234,162 234,169 234,217 234,217	31.79 31.83 31.88 31.91	234,190	3
3. 239,254	239,242 239,291 239,302	100.00 49.32 49.27	239,272	18
4. 240,676	240,654 240,693 240,693	99.98 49.49 49.36	240,674	- 2
5. 241,956	241,927 241,927 241,969	17.61 17.58 35.86	241,948	- 8
6. 243,370	243,324 243,332 243,379	17.51 17.52 35.87	243,352	-18
7. 250,367	250,332 250,383	36.14 35.30	250,358	- 9
8. 251,755	251,740 251,744 251,784 251,784	18.07 18.06 17.71 17.74	251,762	7
9. 253,020	253,018 253,018 253,057	49.34 49.46 99.94	253,036	16
10. 254,425	254,416 254,470	98.51 99.92	254,443	18
11. 259,512	259,495 259,495 259,546	31.85 31.82 63.51	259,521	9
12. 260,934	260,902 260,906 260,946	31.86 31.85 63.65	260,924	-10

<sup>a</sup>Average line error = 25 KHz.

Table A-XIII. Na<sup>23</sup>Br<sup>81</sup> Spectra. V = 1  
 (J=1, M<sub>J</sub>=±1, V=1  $\xrightarrow{E=600 \text{ v/cm}}$  J=1, M<sub>J</sub>=0, V=1)

Observed Line <sup>a</sup> Positions	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
1. 236,706 <sup>b</sup> KHz	236,691 KHz 236,731	63.80 63.86	236,711 KHz	5 KHz
2. 238,121 <sup>b</sup>	238,073 238,080 238,126 238,126	31.81 31.85 31.89 31.92	238,100	-21
3. 243,370 <sup>c</sup>	243,318 243,364 243,375	100.00 49.36 49.32	243,347	-23
4. -- d	244,715 244,752 244,752	99.98 49.52 49.40	244,734	--
5. -- d	246,058 246,058 246,098	17.63 17.60 35.86	246,078	--
6. -- d	247,440 247,447 247,493	17.53 17.55 35.86	247,465	--
7. 254,718	254,667 254,716	36.14 35.36	254,692	-26
8. 256,100	256,060 256,063 256,102 256,102	18.07 18.06 17.73 17.76	256,081	-19
9. 257,443	257,408 257,408 257,445	49.38 49.49 99.93	257,427	-16
10. 258,797 <sup>e</sup>	258,791 258,843	98.60 99.91	258,817	20
11. 264,049	264,034 264,034 264,083	31.86 31.83 63.54	264,059	10
12. 265,464 <sup>f</sup>	265,427 265,430 265,469	31.87 31.85 63.67	265,448	-16

<sup>a</sup>Average line error = 25 KHz.

<sup>b</sup>Overlap with V = 0 Na<sup>23</sup>Br<sup>79</sup> line.

<sup>c</sup>Overlap with V = 0 Na<sup>23</sup>Br<sup>81</sup> line.

<sup>d</sup>Line has not been observed sufficiently well.

<sup>e</sup>Overlap with V = 2 Na<sup>23</sup>Br<sup>79</sup> line.

<sup>f</sup>Overlap with V = 1 Na<sup>23</sup>Br<sup>79</sup> line.



Table A-XIV. Na <sup>23</sup>Br <sup>81</sup> Spectra V = 2  
 (J=1, M<sub>J</sub>=±1, V=2  $\xrightarrow{E=600 \text{ v/cm}}$  J=1, M<sub>J</sub>=0, V=2)

Observed Line Positions <sup>a</sup>	Calculated Line Positions		Composite Line Frequency	Difference
	Calc'd Freq.	Intensity		
1. 240,636 KHz	240,589 KHz 240,628	63.82 63.87	240,609 KHz	-27 KHz
2. 241,956	241,957 241,964 242,007 242,007	31.82 31.86 31.90 31.92	241,981	25
3. 247,322	247,366 247,410 247,834	100.00 49.41 49.37	247,393	71
4. 248,738	248,747 248,783 248,783	99.98 49.55 49.44	248,765	27
5. -- <sup>b</sup>	250,160 250,160 250,198	17.64 17.62 35.85	250,179	
6. -- <sup>b</sup>	251,529 251,535 251,578	17.56 17.57 35.85	251,553	
7. 258,981 <sup>c</sup>	258,974 259,021	36.14 35.41	258,998	17
8. 260,314	260,352 260,355 260,392 260,392	18.07 18.06 17.74 17.78	260,372	58
9. -- <sup>b</sup>	261,769 261,769 261,805	49.40 49.51 99.93	261,787	
10. -- <sup>b</sup>	263,138 263,188	98.68 99.91	263,163	
11. 268,522	268,545 268,545 268,592	31.87 31.84 63.57	268,568	46
12. 270,018	269,923 269,926 269,963	31.87 31.86 63.68	269,943	-75

<sup>a</sup>Average line error = 30 KHz

<sup>b</sup>Line has not been observed inadvertently or sufficiently well.

<sup>c</sup>Overlap with V = 1 Na <sup>23</sup>Br <sup>79</sup> line.

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