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Authors

Hollowell, C.D. Hebert, A.J. Street, J.

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THE MOLECULAR-BEAM ELECTRIC-RESONANCE METHOD*

C. D. Hollowell, A. J. Hebert, and K. Street, Jr.

Lawrence Radiation Laboratory and Department of Chemistry
University of California
Berkeley, California

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ABSTRACT

Radio-frequency transitions of the type $(J, m_J^{\pm}1) \rightarrow (J, m_J)$ have been observed for Na²³F¹⁹ by the molecular beam electric resonance method. Spectra were observed for the first three vibrational states of the J=1 rotational level and for the first vibrational state of the J=2 rotational level. From the analysis of the radio-frequency spectra, dipole moments, μ_V , sodium quadrupole coupling constants, $(eqQ)_V$, spin-rotation interaction constants, c_{Na} and c_F , and spin-spin interaction constants, c_3 and c_4 , were determined for Na²³F¹⁹. Observations of $J=1 \rightarrow J=0$ microwave transitions yielded rotational constants, B_V , for the V=0, 1, and 2 vibrational states. The molecular constants determined for Na²³F¹⁹ are:

$$\mu_{\rm v} = 8.12349 + 0.06436 \, ({\rm v} + 1/2) + 0.0037 \, ({\rm v} + 1/2)^2 \pm 0.001 \, {\rm Debye}$$
 $({\rm eqQ})_0 = -8440.1 \pm 1.5 \, {\rm kc}; \, ({\rm eqQ})_1 = -8327.9 \pm 1.5 \, {\rm kc};$
 $({\rm eqQ})_2 = -8220.5 \pm 2.0 \, {\rm kc}$
 $c_{\rm Na} = 1.4 \pm 0.5 \, {\rm kc}$
 $c_{\rm F} = 2.0 \pm 0.3 \, {\rm kc}$
 $c_{\rm g} = 3.85 \pm 0.25 \, {\rm kc}$
 $c_{\rm g} = 3.85 \pm 0.25 \, {\rm kc}$
 $c_{\rm g} = 0.0 \, \langle {}^{+0.4}_{-0.1} {\rm kc}$
 $c_{\rm g} = 13029.811 \pm 0.002 \, {\rm Mc}; \, B_{\rm g} = 12894.543 \pm 0.003 \, {\rm Mc};$
 $c_{\rm g} = 12760.674 \pm 0.004 \, {\rm Mc}$

I. INTRODUCTION

When the present work was begun, the radio-frequency and micro-wave investigations of NaF by the molecular-beam electric-resonance method had not been reported. However, a microwave study by Bauer and Lew has now been published, and we recently learned of a radio-frequency study by Gräff and Werth. Earlier radio-frequency spectra by the magnetic-resonance method had given somewhat less accurate values for the sodium quadrupole coupling constant than those now reported by the electric-resonance method.

In the present work the molecular-beam electric-resonance method has been used to investigate both the radio-frequency and microwave spectra of $\mathrm{Na}^{23}\mathrm{F}^{19}$. The observed radio-frequency spectra were produced by transitions of the type $(J, m_J^{\pm} 1) \rightarrow (J, m_J)$, where J is the rotational quantum number and m_J is the projection of the rotational angular momentum on the direction of the electric field. The observed microwave transitions were of the type $J = 1 \rightarrow J = 0$.

Values for the electric dipole moments, $\mu_{_{\mbox{V}}}$, sodium quadrupole coupling constants, $(\mbox{eqQ})_{_{\mbox{V}}}$, sodium and fluorine spin-rotation interaction constants, $c_{_{\mbox{Na}}}$ and $c_{_{\mbox{F}}}$, and tensor and scalar spin-spin interaction constants, $c_{_{\mbox{Na}}}$ and $c_{_{\mbox{H}}}$, were determined by analysis of the radio-frequency spectra. The rotational constants, $B_{_{\mbox{V}}}$, were determined from the microwave spectra. Fair agreement was found between the present microwave results and those of Bauer and Lew. 1

II. EXPERIMENTAL

The apparatus and techniques used in these experiments are the same as those used in a previous study of LiBr. 5 As these are described in that report and in more detail elsewhere, 6 only a brief description is given here.

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The apparatus has dipole inhomogeneous electric deflecting fields, a homogeneous electric Stark "C" field region 25 cm long, and a platinum-iridium tube-oven source. Detection is by surface ionization on a tungsten ribbon and mass analysis. Since in the present experiments magnetic effects could be expected to produce noticeable perturbations, care was taken to reduce all components of the earths magnetic field and fringe magnetic fields from the mass spectrometer to less than 50 milligauss along the "C" field transition region.

Radio-frequency transitions of the type $(1, \pm 1) \rightarrow (1, 0)$ and $(2, \pm 2) \rightarrow (2, \pm 1)$ were observed for the lower vibrational states of NaF. The signals observed in the experiments reported here are commonly referred to as "flop-in" signals, since the molecule must undergo a transition to reach the detector. For the J=1 rotational state, the spectrum consisted of six major line groups with hyperfine splittings resolvable in five of the six groups. Figure 1 shows the v=0, J=1 spectrum of NaF at an electric field strength of 250 volts/cm.

The observed line widths were approximately 3 kc at half-maximum intensity for the resolved lines, in agreement with the expected theoretical uncertainty broadening for a 25-cm transition region. The signal-to-noise ratio was approximately 20 to 1 for observed transitions of the v=0 vibrational state.

Also shown in Fig. 1 are the calculated line positions and their relative intensities. Figure 2 shows the v=0, J=0 and 1 energy levels of NaF with respect to the electric field strength and the observed six major radio-frequency transitions in the absence of spin-rotation and spin-spin interaction hyperfine structure.

The $J=1\rightarrow J=0$ microwave transitions were observed by setting the Stark field and the radio frequency to give a maximum signal for a

prominent line in the reorientation spectrum, i.e. a J=1, $m_J=\pm 1 \rightarrow J=1$, $m_J=0$ transition. The microwave frequency was then swept and the resonant frequency observed as a decrease in the radio-frequency "flop-in" signal due to depletion of the final state by rotational transition of the type J=1, $m_J=0 \rightarrow J=0$, $m_J=0$. The decrease amounted to a more than 40% reduction in the "flop-in" Stark signal. The microwave line widths at half maximum were approximately 30 kc.

Figure 2 shows the two major microwave transitions that could be observed for NaF using the above technique. Transitions labelled type II were observed for the first three vibrational states at an electric field strength of 50 V/cm and both type I and II transitions were observed for these vibrational states at a field strength of 400 V/cm.

III. THEORY

The spectra were analyzed with a high speed digital computer program using the Hamiltonian: 7,8

$$\begin{array}{lll}
 & = & B \tilde{\Pi}_{5} - \tilde{\Pi} \cdot \tilde{\Pi}^{5} \cdot \tilde{$$

The first term in the above expression gives the rotational energy, where B is the molecular rotational constant and J is the

rotational angular-momentum operator. The second term gives the interaction of the permanent electric-dipole moment, μ , of the molecule with the external electric field, E The third and fourth terms give the interaction of the nuclear electric-quadrupole moments (Q_1 and Q_2) with the electric-field gradients at the nuclei (\mathbf{q}_1 and \mathbf{q}_2), \mathbf{e}_1 is the electronic charge, I_1 and I_2 are the nuclear spins, and the subscripts distinguish between the two nuclei. The fifth and sixth terms give the magnetic coupling of the nuclear spins with the molecular angular momentum, where c_1 and c_2 are constants. The seventh and eighth terms represent the tensor and scalar spin-spin interactions, 9 respectively. The tensor spin-spin interaction constant, c_3 , is defined as $c_3 = c_{dir}$ + c_{ind} . The direct spin-spin interaction constant, c_{dir} , arises from the usual magnetic dipole-dipole interaction between the two nuclei, and equals $g_1 g_2 \mu_N^2 \langle \frac{1}{3} \rangle$, where g_1 and g_2 are the nuclear g factors for the two nuclei, r is the internuclear distance, and μ_{N} is one nuclear magneton. The indirect spin-spin interaction constant, cind, arises from the tensor component of an electron-coupled nuclear spin-spin interaction. The scalar spin-spin interaction constant, c_{j_1} , arises from the scalar component of an electron-coupled nuclear spin-spin interaction.

In the case of Na 23 F¹⁹, the F¹⁹ nucleus, with spin I = 1/2, has no quadrupole moment. Therefore in the analysis of the radiofrequency spectra, inputs to the program included the two known values, B_v and E, and the unknown parameters, μ_{v} (eq_{Na}Q_{Na})_v, c_{Na}, c_F, c₃, and c₄ for each vibrational state.

The computer program calculated the matrix elements of \mathbb{N} in a J, I_1 , I_2 , m_1 , m_2 , representation, where m_J , m_1 , and m_2 are the projections of \underline{J} , \underline{I}_1 , and \underline{I}_2 respectively on the direction of the

field E. The program computes the energy eigenvalues by diagonalizing this matrix and then calculates the spectral-line positions corresponding to the given set of input parameters according to the selection rules $\Delta m_F = 0, \pm 1 \text{ where } m_F \text{ is the projection of the total angular momentum}$ on the field direction. The unknown parameters are varied to obtain a best fit to the observed spectra.

The quadrupole terms have matrix elements diagonal in J as well as ones connecting J with J ± 2. The quadrupole operator in the Hamiltonian given above can be used only to calculate matrix elements diagonal in J; therefore, in order to include the off-diagonal elements, a more general expression is needed. Formulae derived by Fanoll were used in calculating these matrix elements.

The nonzero matrix elements of the second term in the Hamiltonian, the Stark interaction, are of the form $(J, m_J | \underline{\mu} \cdot \underline{E} | J \pm 1, m_J)$. Since J may take any positive integer value, the matrix is infinite in extent. However, in calculating J = l eigenvalues, only the first four J states are included in the matrix; for J = 2 calculations, the first five J states are included, and so on. For the Stark energy this is equivalent to a fourth-order perturbation treatment, whereas for the quadrupole energy it is equivalent to a second-order perturbation treatment.

IV. RESULTS AND DISCUSSION

(A) Radio-Frequency Spectra

Table I lists the observed and calculated line positions of NaF at electric field strengths of 250 and 500 volts/cm for the J=1 and J=2 rotational states, respectively. In the case of unresolved multiplets, calculated composite line positions were obtained by graphically adding component lines that had the experimental singlet line shapes and calculated relative intensities.

The relative intensities of the lines were obtained using a program 12 which calculates the line strengths from the matrix elements of the dipole moment operator using the computed wave functions. This program uses essentially the same Hamiltonian as discussed above but does not include the off diagonal matrix elements of the quadrupole operator and consequently does not include the Stark-quadrupole interaction and second-order quadrupole effects. The program has given good relative intensities for the spectra of ${\rm Li}^6{\rm F}, {}^{13}, {}^{14}$ and ${\rm Li}^7{\rm F}, {}^{14}$ This calculation was used only for the intensities; the line positions were determined in all cases by the more complete calculation. The maximum difference in any line position on including the Stark-quadrupole and second-order quadrupole effects was just under 2 kc for ${\rm J}=1$ spectra and 5 kc for ${\rm J}=2$ spectra.

Table II lists the dipole moments for the first three vibrational states of NaF As has been observed for LiF¹⁴, 15 and LiBr, 5 a non-linear variation of the dipole moment with vibrational state has been found and the observed dipole moments have been fitted to the expression:

$$\mu_{v} = \mu_{e} + \mu_{I} (v + 1/2) + \mu_{II} (v + 1/2)^{2}$$

The coefficients are also given in Table II. Our dipole moments are significantly lower than those given by Bauer and Lew. 1

The quadrupole coupling constants have also been fitted to an expression quadratic in the vibrational number. The quadrupole coupling constants for the first three vibrational states and the terms of the quadratic expression are shown in Table III.

The coefficient of the quadratic term is required for a best fit, although it is just at the limit of the experimental error and may not be significant.

Our quadrupole coupling constants are in agreement with those of Bauer and Lew within the combined experimental errors.

The sodium and fluorine spin-rotation interaction constants did not show any significant change with vibrational state. The determined spin-rotation constants are:

$$c_{Na} = 1.4 \pm 0.5 \text{ kc}$$

$$c_F = 2.0 \pm 0.3 \text{ kc}$$

In a first attempt to obtain a good fit between the observed and calculated line positions, the value of $c_{\rm dir}$, obtained from the nuclear magnetic moments and $\langle \frac{1}{7} \rangle$, was used as the input for c_3 and only the parameters μ_v , $(eqQ)_v$, $c_{\rm Na}$, $c_{\rm F}$, and c_4 were varied. However, a good fit could not be found until c_3 was also varied with the result that c_3 was determined to be smaller than $c_{\rm dir}$. Our experimentally determined value of c_4 is 3.85 \pm 0.25 kc, while $c_{\rm dir}$ was calculated to be 4.157, 4.101 and 4.046 kc for the v=0, 1, and 2 vibrational states, respectively. No significant change with vibrational state was observed for c_3 .

Large values of c_{l_1} (-11.64 kc) have been reported for TIF. Attempts to extract a nonzero value for c_{l_1} from the data were unsuccessful. Our calculations indicate that c_{l_1} can not be reliably determined with the present line widths (~3 kc) and line position accuracies (~200 cps). However, our observations do allow us to place an upper and a lower limit of + 400 cps and - 100 cps, respectively, on this quantity.

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(B) Microwave Spectra

The observed microwave transition frequencies were corrected for Stark and hyperfine splitting by using the dipole moments and interaction constants determined in the radio-frequency experiments.

Microwave transitions were observed using Stark fields of both 400 V/cm and 50 V/cm. With the stronger field the uncertainty in the Stark correction was approximately 10 kc whereas with the weaker field this uncertainty was well under 1 kc. Thus, although both results agree well within the experimental error, only the more accurate weak-field results are given.

Table IV lists the observed microwave frequencies, the Stark and hyperfine corrections, and the corrected microwave frequencies.

The spectroscopic constants were determined using the familiar Dunham expressions. ¹⁶ The vibrational constants, ω_e and $\omega_e x_e$, as given by Ritchie and Lew¹⁷ for NaF were used to calculate D_e and B_e . Table V lists the spectroscopic constants for Na²³F¹⁹ as determined from the present work along with those given by Bauer and Lew.

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- *This work was supported by the U.S. Atomic Energy Commission.
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Table I. Observed and calculated line positions for the radiofrequency spectra of Na23F19

ne No.	Observed Line Positions (kc)	Calculated Line Positions (kc)	Calculated Relative Line Intensities	Final Calculated Composite Line Positions (kc)
	(1, ±	$(1,0), v = 0, E_c$	= 250 V/cm	
l _a	9909.39 ± .17	9909.30	61	9909.30
1 _b	9913.97 ± .24	9912.84	31	9913.98
- Ъ	9917.9124	9915.12	31∫	9917.90
2 a	10945.08 ± .11	10945.04	100	10945.04
р 2	10953.47 ± .11	10953.42	100	10953.42
3.	11592.12 ± .30	11591.02 11591.15 11593.31 11594.19	18 18 18 18	11592.42
4 a	12921.04 ± .09	12920.77 12921.84	37 18	12921.04
4 _b	12928.55 ± .29	12928.84	18	12928.84
5 _a	13564.69 ± .14	13564.39 13566.21	98 } 49 }	13564.84
5 _b	13569.44 ± .38	13569.24	49	13569.24
6 _a	14601.13 ± .35	14600.03 14602.63	32) 31)	14601.33
6 _b	14606.48 ± .13	14605.66 14607.03	32) 32)	14606.35
	(1, ±	1) \rightarrow (1,0), v = 1, E _c	= 250 V/cm	
l _a	10256.39 ± .17	10256.34	23	10256.34
1 _b	10260.78 ± .30	10259.89 10262.17	11}	10261.03

Table I. Continued

ine No.	Observed Line Positions (kc)	Calculated Line Positions (kc)	Calculated Relative Line Intensities	Final Calculated Composite Line Position (kc)
• •	(1,	\pm 1) \rightarrow (1,0), $v = 1$, E	c = 250 V/cm	
2 _a	11280.32 ± .14	11280.40	37	11280.40
2 _b	11288.85 ± .14	11288.85	37	11288.85
3	11917.19 ± .27	11915.98 11916.13 11918.26 11919.17	7 7 7 7	11917.39
4 a	13229.51 ± .11	13229.19 13230.30	14	13229.47
4 ъ	13237.27 ± .20	13237.30	7	13237.30
5 _a	13862.36 ± .14	13861.75 13863.65	36 18	13862.23
5 _b	13866.77 ± .27	13866.68	18 .	13866.68
6 _a	14887.71 ± .12	14886.39 14888.98	12	14887.69
6 _b	14892.75 ± .17	14892.02 14893.39	12	14892.71
	(1,	\pm 1) \rightarrow (1,0), v = 2, E	= 250 V/cm	
la	10611.71 ± .32	10611.60	9	10611.60
1 _b	10616.08 ± .37	10615.14 10617.38	1 }	10616.26
2 _a	11625.03 ± .19	11625.08	14	11625.08
2 _b	11633.55 ± .20	11633.42	14	11633.42

Table I. Continued .

ine No.	Observed Line Positions (kc)	Calculated Line Positions (kc)	Calculated Relative line Intensities	Final Calculated Composite Line Positions (kc)
	(1, ±	1) \rightarrow (1,0), v = 2, E _c	= 250 V/cm	
3	12251.75 ± .40	12250.20 12250.35 12252.44	3 3 3	12251.59
l _t a	13547.85 ± .16	12253.35 13547.43 13548.56	3 6 3	13547.71
5 _a	14170.18 ± .23	14169.90 14171.79	14	14170.37
6 _b	15189.79 ± .23	15189.18 15190.54	5)	15189.86
	(2, ±	2) \rightarrow (2, \pm 1), $v = 0$,	$E_c = 500 \text{ V/cm}$	
2 _a	10734.62 ± .08	10734.70	100	10734.70
2 _b	10740.86 ± .08	10740.86	100	10740.86
3 _a	11134.86 ± .24	11134.89	32	11134.89
3 _b	11139.07 ± .29	11138.74 11139.69	14 14	11139.21
⁵ a ⁵ ზ	12484.89 ± .15 12488.27 ± .21	12484.90 12488.26	95 95	12484.90 12488.26

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Table II. Dipole moments for Na²³F¹⁹

$$\mu_{0} = 8.15576 \pm 0.001 D^{8}$$
 $\mu_{e} = 8.12349 \pm 0.0015 D$
 $\mu_{l} = 8.22086 \pm 0.001 D$
 $\mu_{l} = 0.06436 \pm 0.0008 D$
 $\mu_{l} = 0.00037 \pm 0.0003 D$
 $\mu_{v} = \mu_{e} + \mu_{l} (v + 1/2) + \mu_{l} (v + 1/2)^{2}$

The accuracy of the dipole moments (± 0.001 D) is limited by the uncertainty in the absolute value of the electric Stark field, however, the precision of the results is at least an order of magnitude better (± 0.0001 D). See Ref. 5.

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Table III. Sodium quadrupole coupling constants for Na 23 F19.

$$(eqQ)_0 = -8440.1 \pm 1.5 \text{ kc}$$
 $(eqQ)_e = -8498.0 \pm 2.6 \text{ kc}$
 $(eqQ)_1 = -8327.9 \pm 1.5 \text{ kc}$ $(eqQ)_1 = +117.0 \pm 4.1 \text{ kc}$
 $(eqQ)_2 = -8220.5 \pm 2 \text{ kc}$ $(eqQ)_{II} = -2.4 \pm 2.5 \text{ kc}$
 $(eqQ)_v = (eqQ)_e + (eqQ)_I (v + 1/2) + (eqQ)_{II} (v + 1/2)^2$

1

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Table IV. Observed microwave transition frequencies and Stark—hyperfine corrections. All transitions were observed at $E_{\rm c}=50.0$ V/cm.

Vibrational State	Line Number	Observed Transition Frequencies (Mc/sec)	J = 0 and J = 1 Stark and Hyperfine Corrections (Mc/sec)	Corrected Transition Frequencies (Mc/sec)
v = 0	II	26062.1602 ± 0.0025	2.6771 ± 0.0005	26059.4831 ± 0.0030
v = 1	11	25791.6142 ± 0.0035	2.6657 ± 0.0005	25688.9485 ± 0.0040
v = 2	II	25523.8680 ± 0.0050	2.6560 ± 0.0006	25521.2120 ± 0.0056

Table V. Spectroscopic Constants for Na 23F19.

	Pre	esent Resu	lts	Bauer	and Lew ^a	
D _e (~ -Y ₀₂)	0.0347	± 0.0003	Мс	0.0347	± 0.0003	Mc
β _e (~ -Υ ₁₂)	- 0.00024	± 0.0000	+ Mc		• • • •	ر ماران ماران
Yol (~B _e)	13097.970	± 0.005	Mc	13097.971	± 0.003	Mc
$Y_{11} (-\alpha_{e})$	- 136.667	± 0.007	Me ·	- 136.665	± 0.004	Ме
Υ ₂₁ (~γ _e)	0.6995	± 0.004	Mc	0.700	± 0.001	Мс
Be	13098.029	± 0.060	Mc	13098.032	± 0.035	Мс
$^{\mathrm{B}}\mathrm{O}$	13029.811	± 0.002	Mc	13029.813	± 0.001	Mcb
B	12894.543	± 0.003	Me	12894.548	± 0.001	Me
B ₂	12760.674	± 0.004	Me	12760.683	± 0.001	Mcb
	$B_v = Y_{Ol} + Y_{ll}$	(v + 1/2)	+ Y ₂₁ ($(v + 1/2)^2$		

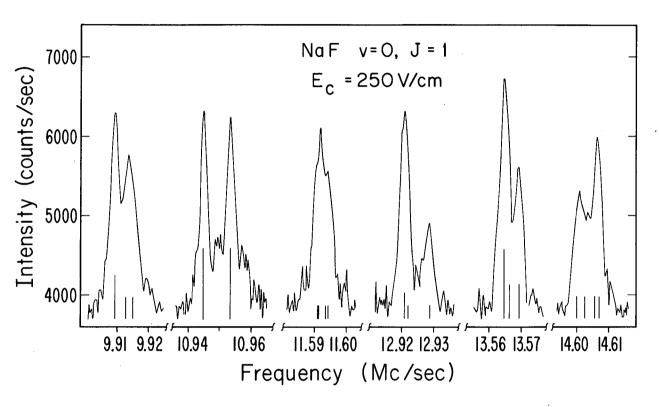
aRef. 1.

^bIt is not clear to us whether the uncertainties given here represent the total expected error or simply the precision of measurement of the observed frequencies.

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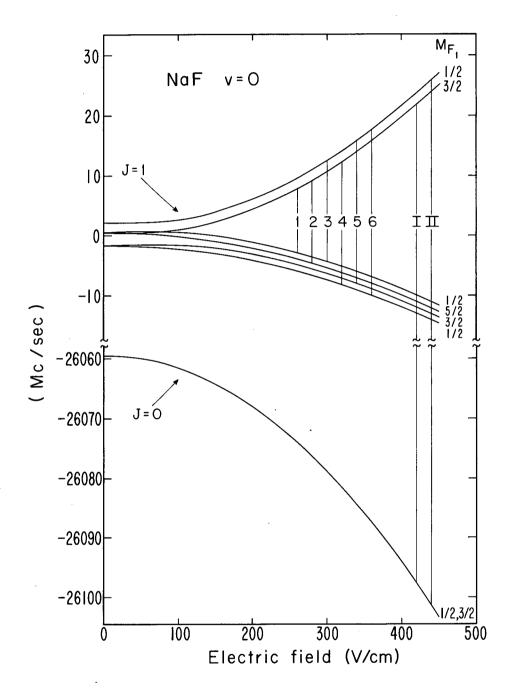
FIGURE CAPTIONS

- Figure 1. A radio-frequency spectrum of Na $^{23}F^{19}$ in the v=0 vibrational state. Transitions of the type $(1, \pm 1) \rightarrow (1,0)$ were observed at an electric field strength of 250 V/cm.
- Figure 2. The energy levels of $Na^{23}F^{19}$ with respect to the electric field strength. Arabic numerals refer to observed radio-frequency transitions and Roman numerals refer to microwave transitions. $M_{F_1} = m_J + m_{T_1}$.



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Fig. 1



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Fig. 2

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