the data of Fig. 5 for each value of the energy transfer, ignoring the extrema in the structure. The zero-order terms were then used to correct the scattering law data and re-evaluate the first moment $\langle\epsilon^1\rangle$ which is displayed in Fig. 7. The corrected first moment agrees much better with theory; however, the procedure used in correcting the data is open to question considering the fact that beryllium is a coherent scatterer.

V. CONCLUSIONS

The scattering law has been measured for a powdered beryllium sample at room temperature for energy transfers up to and including $\epsilon=4.0$ ($\hbar\omega=0.1027$ eV). Observed structure in the scattering law data shows that the interference condition for neutron-phonon interactions in coherent scatterers manifests itself in the details of the scattering from even a polycrystal sample, and that the structure is most pronounced at low frequencies (low-energy transfers) where the conditions for scattering are most restrictive. It would appear from this experiment that the incoherent approximation is inadequate for describing the details in the scattering data. The first moment of the energy transfer with respect to the scattering law has been evaluated for these data and found to be high at low-momentum transfers. This discrepancy is attributed to multiple scattering in the sample.

ACKNOWLEDGMENTS

The author is indebted to Dr. R. M. Brugger and Dr. P. D. Randolph for helpful discussions regarding the interpretation of the results of the experiment, to Miss B. K. Sidle for handling the data reduction and programming the averaging procedure, and to Dr. R. G. Fluharty for continued support.

PHYSICAL REVIEW VOLUME 136, NUMBER 5A 30 NOVEMBER 1964

Hyperfine Structure of V_k Centers in Alkali Fluoride Crystals*†

C. EDWARD BAILEY
Department of Physics, Dartmouth College, Hanover, New Hampshire

(Received 2 July 1964)

V_k centers in LiF, NaF, KF, RbF, and CsF are studied. It is shown that the V_k center is stable in all the alkali fluorides at liquid-nitrogen temperature and unstable at room temperature. The results for LiF are in excellent agreement with earlier results. The predictions of the theoretical Hamiltonian derived by Castner, Woodruff, and Känzig are in excellent agreement with all of the experimental results. Values of $g_{\parallel}$, $g_{\perp}$, a, and b are presented for NaF and KF. It is shown that the linewidths of the V_k spectrum depend rather heavily on the nearest-neighbor atoms (the alkalis), and the relaxation process seems to be one of hyperfine coupling.

I. INTRODUCTION

MANY different color centers have been studied in the alkali halides.[1] We will treat the V_k center in the alkali fluorides in some detail. This center has been studied in great detail by Castner, Woodruff, and Känzig.[2-4] The electron paramagnetic resonance (EPR) data have shown that this center is a hole shared between two negative halide ions in the crystal or, in other words, can be thought of as the molecular-ion X_2^-. Optical studies are in agreement with this model.[5-7] Gassinelli and Mieher, using electron-nuclear double-resonance (ENDOR) techniques on LiF, show that their data can be explained by this model.[8,9] From this model it is seen that the centers can lie along each of the six equivalent $\langle 110\rangle$ directions and in the absence of strain or other anisotropic conditions will populate these sites with equal abundance.

The EPR spectrum that one observes for this center results from a very large electron-nuclear interaction, giving rise to a well-resolved hfs. Because this is a hole center, the wave function of the unpaired hole may be

---

* This work was supported by a grant from the National Science Foundation.

† Based on a thesis submitted to the faculty of Dartmouth College in partial fulfillment of the requirements for the M.A. degree.

[1] See, for example, F. Seitz, Rev. Mod. Phys. **26**, 7 (1956); or J. H. Schulman and W. D. Compton, *Color Centers in Solids* (Pergamon Press, Inc., New York, 1963).

[2] W. Känzig, Phys. Rev. **99**, 1890 (1955).

[3] T. G. Castner and W. Känzig, Phys. Chem. Solids **3**, 178 (1957).

[4] T. O. Woodruff and W. Känzig, Phys. Chem. Solids **5**, 268 (1958).

[5] C. J. Delbecq, B. Smaller, and P. H. Yuster, Phys. Rev. **111**, 1235 (1958).

[6] C. J. Delbecq, W. Hayes, and P. H. Yuster, Phys. Rev. **121**, 1043 (1961).

[7] W. Hayes and G. M. Nichols, Phys. Rev. **117**, 993 (1960).

[8] R. Mieher and R. Gazzinelli, Bull. Am. Phys. Soc. **9**, 88 (1964).

[9] R. Gazzinelli and R. Mieher, Phys. Rev. Letters **12**, 644 (1964).

expected to be rather compact in contrast with that of the unpaired electron in the F center. This results in the hole interacting mainly with the two adjacent halide nuclei and with the other neighboring atoms to a much lesser extent. To study the influence of the surrounding ions on the hfs of the V_k center, it is desirable to examine the same center X_2^- in a number of crystals; that is, to keep the halide fixed and vary the alkali ions. The alkali fluorides are singled out because the fluorine atom has a nuclear spin I equal to one-half, a large magnetic moment, and only one isotope. This causes the spectrum for the F_2^- molecular ion to be especially simple and the separate lines well resolved.

The V_k center in LiF has been studied in great detail by Castner, Woodruff, and Känzig.[3,4] Phelps[10] has made a study of V_k centers in NaF and has found that the hfs is qualitatively similar to LiF. We present here a study of the hyperfine interaction of the V_k center in LiF, NaF, KF, RbF, and CsF.

II. THEORETICAL MODEL

Woodruff and Känzig[4] show that the Hamiltonian for the V_k center takes the form

$$\mathcal{H}=g_{\parallel}\beta_0 H_{z'}S_{z'}+g_{\perp}\beta_0(H_{x'}S_{x'}+H_{y'}S_{y'}) +g_0\beta_0(a\mathbf{I}\cdot\mathbf{S}+bS_{z'}I_{z'})+g_N\beta_N\mathbf{H}\cdot\mathbf{I}, \quad (1)$$

with

$$a=\mu_I[\tfrac{4}{3}\pi\beta^2|\Psi(0)|^2-\tfrac{1}{5}\alpha^2\langle r^{-3}\rangle_{\mathrm{av}}] \quad (2)$$

and

$$b=\mu_I[\tfrac{3}{5}\alpha^2\langle r^{-3}\rangle_{\mathrm{av}}], \quad (3)$$

where μ_I is the magnetic moment of the halide nucleus, β^2 and α^2 represent the fraction of s and p states, respectively, $|\Psi(0)|$ is the amplitude of the s function at the nucleus, and $\langle r^{-3}\rangle_{\mathrm{av}}$ has to be taken over the p function centered on the nucleus. The approximation of cylindrical symmetry has been made so that $g_{\parallel}$ is the g value along the molecular bond axis and $g_{\perp}$ is the value perpendicular to this axis. The z' refers to an axis that is along the molecular bond axis. They solve Eqs. (2) and (3) for β^2 and get

$$\beta^2=\frac{(a/b+\frac{1}{3})}{(a/b+\frac{1}{3})+(20\pi|\Psi(0)|^2)/(9\langle r^{-3}\rangle_{\mathrm{av}})}. \quad (4)$$

Following Castner and Känzig[3] we have used the values[11,3]
$\langle r^{-3}\rangle_{\mathrm{av}}=60.0\times10^{24}\ \mathrm{cm}^{-3}$ and $|\Psi(0)|^2=3.32\times10^{24}\ \mathrm{cm}^{-3}$ in calculations for the amount of s state that are made later.

If the approximation of cylindrical symmetry is made, the equations for the g values derived by Inui, Harasawa, and Obata[12] become

$$g_{\parallel}-g_0\approx-\alpha^2\lambda^2/\Delta^2, \quad (5)$$

$$g_{\perp}-g_0\approx(-2\alpha^2\lambda/\Delta)-(2\alpha^2\lambda^2/\Delta^2), \quad (6)$$

where Δ is the energy level spacing between the $np\sigma_u$ and $np\pi_u$ levels ($n=2$ in the case of fluorine), and λ is the spin-orbit coupling constant.

Woodruff and Känzig[4] show that using a complete set of eight eigenfunctions, an 8×8 matrix can be written from the matrix elements of the Hamiltonian. Although we will use this matrix in the calculations made later, it will not be repeated here. (The angle ϕ of their matrix was set equal to zero in our calculations.)

III. EXPERIMENTAL METHODS

Samples of about 5×12×2 mm were placed in the bottom of a special microwave cavity. Two different orientations of the crystal in the cavity were required in this experiment to permit observation of spectra with the external magnetic field along the [100], [110], and [111] crystalline axes (called the [100], [110], and [111] spectra below). In one orientation the crystal was placed on the bottom of the cavity. In the other orientation the crystal was placed on a Styrofoam wedge at an angle of 45° relative to the cavity bottom. The Styrofoam did not exhibit any paramagnetic centers after x-raying.

Since the V_k centers are unstable at room temperature it is necessary to produce them by x-raying at liquid-nitrogen temperature, and to observe the EPR spectrum without allowing the sample to warm up. The cavity has an aluminum foil bottom to admit the x rays. After the cavity had been attached to a stainless steel cavity arm, the sample in the cavity was then x-rayed from below in a Dewar system through a mica x-ray port[10] for the desired length of time (4–16 h) with a Norelco high-intensity source at 50 kV and 40 mA. Because of the large heat capacity of the cavity, the samples did not warm up appreciably when the cavity and arm were quickly switched to a double Dewar system placed between the pole faces of the magnet of a spectrometer. This X-band superheterodyne spectrometer has been described elsewhere.[13] The magnetic field was measured by proton resonance techniques. The frequency at which the proton resonance occurred and the klystron frequency in the EPR spectrometer were measured to about 1 part in 10^5 with frequency meters calibrated against WWV.

IV. EXPERIMENTAL RESULTS

All of the alkali fluorides were examined, and in every case EPR spectra due to the V_k center, and similar to those seen by Woodruff and Känzig[4] in LiF, were observed. It was found that as the alkalis got heavier the intensity of the EPR spectrum for a given x-raying time decreased. Even after x-raying for 16 h the V_k spectrum in CsF was so weak that it was not possible to get reliable quantitative data from this sample. In many of the samples centers other than the V_k center

---

[10] D. H. Phelps, M. A. thesis, Dartmouth College, 1962 (unpublished).

[11] R. Sternheimer, Phys. Rev. **84**, 244 (1951).

[12] T. Inui, S. Harasawa, and Y. Obata, J. Phys. Soc. Japan **11**, 612 (1956).

[13] W. T. Doyle, Phys. Rev. **126**, 1421 (1962).

were seen. These centers could usually be identified (e.g., U_2 and F centers). Fortunately the physical properties of the V_k center differed from those of the other centers. In no case, except RbF, did these other centers cause serious difficulty. When there was a doubt to which spectrum a particular line belonged, saturation and stability measurements permitted easy recognition of the V_k centers.

The LiF crystals were obtained from Harshaw. After x raying for 8 h the signal was so strong that no noise could be seen on the chart recorder when tracing out the spectrum. The data obtained for LiF will be seen to be in excellent agreement with the results of Känzig and Woodruff.[4]

In the case of NaF both pure crystals from Harshaw and crystals doped with HF (grown in this laboratory) were used. After four hours of x-raying, the signal from the doped sample was greater than that observed in the Harshaw crystals after a 10-h x-ray dose. The mechanism for the greater production rate in the doped samples has been investigated by Doyle and Phelps.[14] Several investigators have studied the effect of doping and found no effect on the EPR spectra.[3,5,6] On the other hand, Hayes and Nichols,[7] who worked with crystals doped with divalent atoms, found that there was a change in the EPR spectrum due to the formation of V_k centers at the divalent impurity site. No production of V_k centers at special sites was observed in our doped samples.

The crystals of KF were obtained from two companies, Harshaw and Light & Company. The KF samples were x-rayed for about 10 h. This gave a good signal-to-noise ratio, but was not as large as that for either LiF or doped NaF.

The sample of RbF from Harshaw after being x-rayed for 12 h gave a signal that was usable, but weak. There seemed to be an impurity line in the center of the spectrum which partially obscured the lines needed to obtain accurate g values. The g values reported for RbF must thus be considered tentative. However, the g values obtained do appear to follow the general trends.

The crystal of CsF from Harshaw was x-rayed for 16 h and the signal was still too small to get any usable quantitative data. The V_k spectrum, however, was identified by the characteristic lines in the [100] spectrum.

hfs of the V_k Center

Figure 1 shows the actual spectrum seen for NaF when the magnetic field is along the [100], [110], and [111] directions. The series of equally spaced additional peaks in the center of the [100] spectrum is due to a small concentration of F centers. This is an undoped sample. The [110] and [111] spectra were from doped samples and it can be seen that the F-center spectrum is greatly reduced. Since the KF record is very similar to this, except that the center of the spectrum is better resolved (because the linewidths are narrower), a copy of this spectrum is not included. Figure 2 shows the line position for LiF as a function of the angle Θ between the [100] crystal direction and the dc magnetic field. It should be noted that the most convenient angle experimentally is the angle Θ. The significant angle for each center is the angle θ between the molecular axis and the external field. Graphs of line position versus θ are not included for the various samples since, except on a very large scale, they look the same as that given by Woodruff and Känzig[4] for LiF. In general, the spectrum observed will contain four sets of lines corresponding to the angles θ_1, θ_2, θ_3, and θ_4 (notation of Woodruff and Känzig[4] has been used).

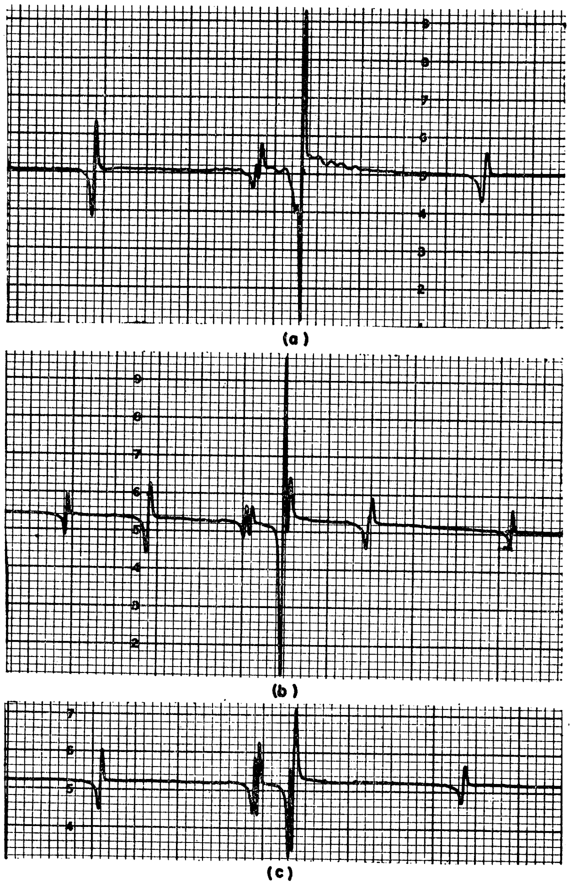

FIG. 1. Spectra for NaF. (a) [100] spectrum, (b) [110] spectrum, and (c) [111] spectrum.

At certain Θ's some of the θ_i's become equal. This is true along the [100], [110], and [111] directions and it is seen in these cases that the spectra are greatly simplified as compared with those in an arbitrary direction. Because of this, careful field positions of all the lines in these special directions were taken. From

---

[14] W. T. Doyle and D. H. Phelps, Program of the International Symposium on Color Centers in Alkali Halides II. Physikalishes Institut der Technischen Hochschule, Stuttgart, Germany, 1962, paper F1, p. 27 (unpublished).

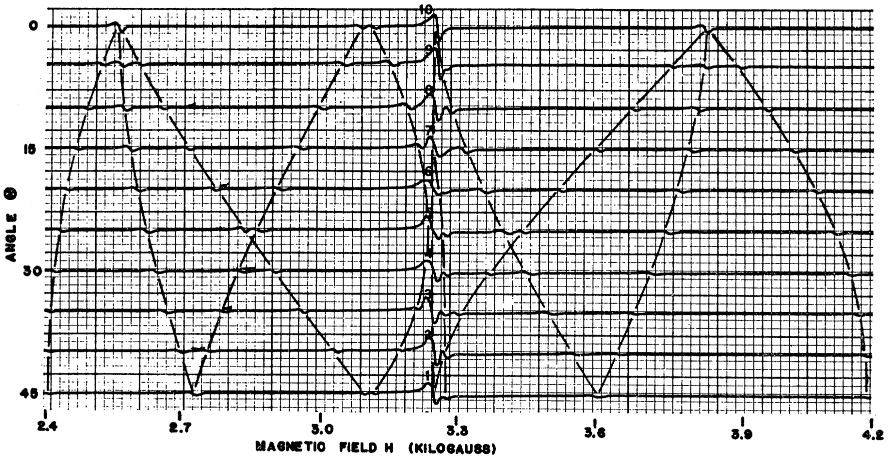

FIG. 2. Observed angular dependence of the line position in LiF.

these readings $a+b$, $g_{\parallel}$, $g_{\perp}$, and $|a|$ were determined after the manner of Woodruff and Känzig[4]; that is,

(1) $g_{\parallel}$ was obtained from R_3 at $\theta=0°$([110] spectrum),
(2) $g_{\perp}$ from R_3 at $\theta=90°$ ([111] spectrum),
(3) $a+b$ from the spacing of R_1 and R_4 at $\theta=0°$, and
(4) $|a|$ (absolute value because the matrix is insensitive to the sign of a) from the position of R_1 at $\theta=90°$.

The constants $g_{\parallel}$, $g_{\perp}$, and $a+b$ are easily obtained directly from the data, while the $|a|$ had to be determined by a numerical fitting process. A simple computer program was used to perform this fitting. The same program was used to calculate theoretical line positions once all of the constants were found.

The quantum of energy δ for each transition remains constant, and the different resonances are obtained by changing H, the magnetic field. The value of $|a|$ was determined as follows: (1) the values of $g_{\parallel}$, $g_{\perp}$, and $a+b$ obtained directly from the data were placed in the computer program, (2) the observed value of the magnetic field for the transition $R_1(90°)$ was used for the value of H in the program, and (3) estimated values of $|a|$ were tried until the experimental δ of the transitions (as obtained by the computer) agreed with that fixed by the klystron frequency. Actually either the value of $R_1(90°)$ or $R_4(90°)$ could have been used, but $R_1(90°)$ was used to follow the procedure of Woodruff and Känzig.[4] In the case of LiF alternate procedures were also tried; that is, simultaneous fitting of R_1 and R_4 by minimizing the deviation of both lines gave a value of $|a|=59$. This small change in $|a|$ (2 G) only affected the 90° spectrum and that not a significant amount. Thus we fit $|a|$ to R_1 and use R_4 as a check.

Table I gives a summary of the numerical results obtained as well as the constants used in the computations. The results given in this table, and elsewhere in the paper, are written to an accuracy indicated by the precision of the measurements and by the spread in values among the different samples used. The error is considered to be in the last figure quoted.

TABLE I. Data and constants. The data for RbF included in brackets are considered to be less certain (see text). The linewidths are computed from the line $R_4(50°)$. β^2 is computed from Eq. (4). The values of Δ/λ and α^2 are obtained by simultaneous solution of Eqs. (5) and (6).

	LiF	NaF	KF	RbF
$a+b$	883.7	897.1	908.0	908.4
$g_{\parallel}$	2.0034	2.0014	2.0020	[2.0034]
$g_{\perp}$	2.0239	2.0220	2.0214	[2.0160]
$\|a\|$	57	47	30	...
b $a>0$	824.7	850.1	878.0	...
b $a<0$	942.7	944.1	938.0	...
Linewidth	12.9	8.9	2.6	11.0
β^2 $a>0$	.51	.50	0.49	...
β^2 $a<0$	.41	.42	0.44	...
Δ/λ	8.8	−12.0	−32.8	...
α^2	.09	.13	0.32	...
ν_e	8775.7	8922.9	9132.3	9171.3
δ	3132.0	3184.5	3259.2	3272.4
β_0	0.92732×10^{-20} ergG^{-1}			
β_N	$\beta_0/1836.14$			
g_0	2.00229			
g_N	2.628			

Having now obtained the values of $g_{\parallel}$, $g_{\perp}$, $a+b$, and $|a|$, values of H at different angles were computed in the same manner as was the $|a|$. As an example of these calculations Table II gives a comparison between experimental and calculated values for KF. Equally

TABLE II. Comparison between calculated and experiment values for KF.

		0°	35.2°	60°	90°
R_1	Exp.	2351.4	2468.6	2691.0	3094.1
	Cal.	2351.4	2467.4	2690.3	3094.1[a]
R_2	Exp.	3258.9	3247.7	3234.5	3099.9
	Cal.	3258.7	3248.4	3235.3	3099.6
R_3	Exp.	3258.9	3247.7	3234.5	3227.7
	Cal.	3258.9[a]	3248.4	3235.4	3227.7[a]
R_4	Exp.	4167.6	3941.4	3587.3	3233.6
	Cal.	4167.6	3946.0	3591.7	3232.4

[a] Fitted.

good agreement was found between the experimental and calculated values of line position for both LiF and NaF.

V. DISCUSSION

The values obtained for the various parameters in LiF are in excellent agreement with those given by Woodruff, Castner, and Känzig.[3,4]

Phelps[10] has studied V_k centers in NaF and found the V_k center in NaF to be qualitatively similar to that in LiF. The data in Table I shows that the V_k centers in all the alkali fluorides are remarkably alike. This general similarity was expected from the similarity found in the Cl_2^- V_k centers in KCl and NaCl.[3] Clearly the F_2^- V_k centers in all of the fluorides conform very well to an "imbedded molecule" picture. The surrounding lattice thus does not have much effect on this center. Although the quantitative results vary very little from sample to sample, certain trends can be seen and these will now be discussed.

Linewidths

As can be seen in Table I the width of the individual lines of the spectra vary from one crystal to the next. This can be attributed to inhomogeneous broadening[15,16] of the lines due to hyperfine (hf) interactions with the surrounding nuclei. Since the nearest neighbors were expected to be most effective in broadening the lines, a plot of linewidth versus the magnetic moment of the alkali was made. See Fig. 3. As this is a straight line that passes fairly close to the origin, it would seem to indicate that the linewidth of the V_k center mainly arises from a hyperfine interaction between the center and its nearest neighbors (the alkalis). This apparently can not be attributed primarily to an effect of the changing ionic radii or interionic distance since there is no correlation between these and the magnetic moments of the alkalis in the different alkali fluorides. The observation that the linewidths depend more heavily on the alkalis than the halides is also supported by the fact that the ratio of linewidths for NaCl and KCl, measured by Castner and Känzig,[3] is equal to the ratio of linewidths for NaF and KF to within the limits of experimental error. The fact that the curve does not pass through the origin is expected as the hf interactions due to the fluorines contribute something to the linewidth. Moreover, the individual spin packets themselves have a natural linewidth due to their finite relaxation times. As the linewidth at the origin is only about 1G, these last effects appear to be small. There seems to be a discrepancy between these observations and the recent ENDOR study on LiF by Gazzinelli and Mieher[8,9] which indicates that the fluorine nucleus ought to play a much more important part in the linewidths. The cause of this discrepancy is not understood at this time.

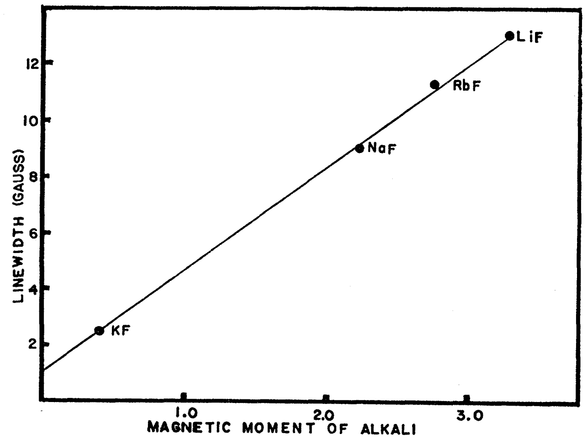

FIG. 3. Dependence of the linewidth on the magnetic moment of the alkali.

Saturation

Arranged in descending order of difficulty to saturate (the most difficult being LiF), the crystals are LiF, RbF, NaF, and KF. It should be noted that this is the same order as for decreasing linewidths. Castner[15] explains that a spin-spin coupling can not account for a relaxation time that depends upon the magnetic field and angle θ. Instead he states that this effect might be explained by a spin-orbit or a hyperfine coupling. A spin-orbit coupling would be expected to depend upon the atomic number of the nuclei and not on the magnetic moments of the alkali. Thus the correlation between linewidth and saturation is consistent with a hyperfine relaxation mechanism.

Hyperfine Interactions

As mentioned previously the matrix can only give the $|a|$. In fact the sign of a could not be determined from our studies of any one crystal. From Table I it is seen that $|a|$ is decreasing as the alkalis increase in atomic weight. This indicates that the amount of s state is either increasing or decreasing depending upon the sign assumed for a. (See β^2 of Table I.) The g shifts indicate that the amount of p state is increasing (see α^2 of Table I). This trend means that the sign of a must be positive. This is in agreement with the sign tentatively assigned from theoretical calculations by Castner and Känzig[3] for NaCl, KCl, and KBr individually.

Das, Jette, and Knox[17] derived the following theoretical equation for computing a:

$$a = a_F - b/3,$$

where a_F is very heavily dependent on the overlap integrals between the two fluorine ions of F_2^- molecular

---

[15] A. M. Portis, Phys. Rev. **91**, 1071 (1953).

[16] T. G. Castner, Phys. Rev. **115**, 1506 (1959).

[17] T. P. Das, A. N. Jette, and R. S. Knox, Phys. Rev. **134**, A1079 (1964).

ion. This means that if a is positive and decreasing as the atomic number of the alkali increases, a_F must be decreasing. This would indicate that the overlap integrals are decreasing. An increase of distance between the two F^- ions in the F_2^- molecular ion would cause this. Das, Jette, and Knox[17] find, by semiphenomenological methods, that the distance between the atoms A and C decreases while that between A and E increases when a F_2^- molecular ion is formed. The notation is as follows: The two atoms A refer to the two fluorine ions in the V_k center, the two atoms C are the nearest alkali atoms (C and D in notation of Das *et al.*), and the four atoms E are the next-nearest alkali atoms (E, F, G, and H in the notation of Das *et al.*). It would seem that if the ionic radii of C (the nearest alkali atoms) are increasing (as is true for alkalis with larger atomic number), the two fluorine ions would be forced further apart. If the sign of a is chosen to be negative, similar argument would indicate that the fluorine ions would be pulled together as the radii of the nearest atoms (C's) increased. Although an elaborate analysis would be required before a positive statement about which sign should be assigned to a can be made, this last information along with that from the g values makes the positive sign seem more likely. Thus the sign of a will be assumed to be positive for the rest of this paper unless specified otherwise.

From Table I it is seen that b changes by only 6.3% while a changes by 47% in the alkali fluorides studied in detail. This is in agreement with the expression for b derived by Das, Jette, and Knox[16] who show the b is not particularly sensitive to R, the spacing between the two ions in the F_2^- molecular ion, while a is quite sensitive to R.

The value of $a+b$ seems to be increasing as the alkali radii are increased, but from a plot of $a+b$ versus interionic spacing it would appear that the value of $a+b$ for CsF would not be greatly different from RbF.

Summary

This study has shown that the V_k center is stable in all the alkali fluorides at liquid-nitrogen temperature and unstable at room temperature; the theoretical picture of the V_k center as derived by Woodruff and Känzig[4] gives a good description of the V_k center in the samples studied in detail; the linewidths depend upon the magnetic moment of the alkali; the sign of a appears to be positive, the amount of p state is increasing; and many of the effects can be explained if the distance between the two ions in the F_2^- molecular ion is increasing as the radius of the alkali ion is increased.

ACKNOWLEDGMENTS

I wish to thank Professor William T. Doyle for suggesting this experiment and for many enlightening discussions and useful suggestions during the course of this work. I also wish to thank A. Agarwal and N. K. Winsor for the writing of the computer program.

PHYSICAL REVIEW VOLUME 136, NUMBER 5A 30 NOVEMBER 1964

Excitation of Coupled Nuclear Antiferromagnetic Resonance Modes*

G. L. Witt† and A. M. Portis
Department of Physics, University of California, Berkeley, California

(Received 19 June 1964)

The presence of a strong hyperfine coupling in the antiferromagnets $KMnF_3$ and $CsMnF_3$ leads to a strong intermixing of the nuclear magnetic resonance and antiferromagnetic resonance (AFR) modes at low temperatures. In the experiments described here, Mn^{55} resonance absorption is detected by monitoring the field for AFR while driving at appropriate nuclear frequencies. Such resonance absorption is observed for a range of frequencies many times larger than the nuclear linewidth. A spin-pinning model is proposed which provides nucleation centers for this "off-resonance" saturation.

1. INTRODUCTION

In a previous paper[1] it was shown that the presence of a large hyperfine coupling in antiferromagnetic $KMnF_3$ leads to a substantial mixing of the nuclear and electronic resonance modes. The results reported there are in agreement with the theoretical calculations of deGennes *et al.*[2] who demonstrated that these effects can be thought of as arising from the coupling of the nuclei through the Suhl-Nakamura indirect interaction.[3] As a consequence, it is found that the field for antiferromagnetic resonance (AFR) and the nuclear magnetic resonance (NMR) frequency are depressed at lower temperatures.

---

* Supported by the U. S. Atomic Energy Commission.

† National Science Foundation Cooperative Fellow for the period during which this work was performed.

[1] G. L. Witt and A. M. Portis, Phys. Rev. **135**, A1616 (1964).

[2] P. G. deGennes, P. A. Pincus, F. Hartmann-Boutron, and J. M. Winter, Phys. Rev. **129**, 1105 (1963).

[3] H. Suhl, Phys. Rev. **109**, 606 (1958); T. Nakamura, Progr. Theoret. Phys. (Kyoto) **20**, 542 (1958).