

Paramagnetic Resonance Hyperfine Structure of V^{50} and V^{51} †

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The paramagnetic resonance has been observed of a magnetically dilute crystal of vanadium Tutton salt containing 23 percent of the isotope V^{50} . The hfs of V^{50} was completely resolved, enabling us to conclude that its spin $I=6$. The ratio of the nuclear g factors, $g(V^{50})/g(V^{51})$, was measured with the result 0.3792 ± 0.0008 , in good agreement with the value determined directly by nuclear magnetic resonance experiments.

THE paramagnetic resonance spectrum of the most abundant isotope of vanadium V^{51} has been investigated by Bleaney, Ingram, and Scovil,¹ and also by Hutchison and Singer.² They established unambiguously the value $7/2$ for the nuclear spin of V^{51} , in contrast to the optical hfs measurements which gave this result with less certainty. It was anticipated that the method of paramagnetic resonance would be applicable to V^{50} which was expected, on the basis of the shell model, to have a spin possibly as great as 7. The principal results of this investigation are an unambiguous determination of the spin of V^{50} , $I=6$,³ and an evaluation of the magnitude of the ratio of the g factors of the two isotopes in excellent agreement with that obtained from nuclear resonance studies.

1. THEORY

The phenomenon of hfs of paramagnetic ions in crystals was observed first by Penrose.⁴ Subsequently Bleaney and his co-workers have successfully applied this method to check earlier spin measurements and to determine the spins of a number of rare earth nuclei.⁵ The general principle involved might be called the Back-Goudsmit effect in solids,⁶ and its theory can be understood qualitatively by semiclassical arguments. Consider an unpaired electron of a paramagnetic ion. The total field seen by this electron depends, in addition to the externally applied magnetic field, upon the internal magnetic field due to the nucleus. Consequently, for a given external field, there will be $2I+1$ values of the local field corresponding to the $2I+1$ possible orientations of the nucleus. If an rf field is applied perpendicular to the steady field, a net absorption of energy will occur at the Larmor frequency, $\nu = (g\beta/h)(H_{\text{ext}} + H_{\text{nuc}})m_I$. Then, if the external field is held constant,

resonance absorption is expected for $2I+1$ different frequencies. If, on the other hand, the frequency is held constant, as is the case in practice, resonance absorption will occur for $2I+1$ different values of the external magnetic field. Furthermore, since only the electron under-goes transition, all hfs components for a given electron transition will have the same intensity. Thus for spin determination it is necessary merely to count the total number of equally intense and (almost) equally spaced lines.

With slight modifications, the above arguments can be applied to divalent vanadium. The ground state of the free V^{++} ion is $4F_{3/2}$. In the presence of the crystalline electric field the orbital degeneracy of the state is partially removed, the lowest state being nondegenerate in the orbital coordinates. The ground state can then be roughly described as that of an ion with orbital angular momentum zero and spin $S=3/2$. The spin-orbit coupling now produces a further splitting of the degenerate spin levels, so that there results a pair of doubly degenerate levels separated by 0.3 cm^{-1} . An external magnetic field completely removes the degeneracy of the levels. Though in fields corresponding to Zeeman splittings of the order of the crystalline field splittings the spectrum would be quite complex, in fields of the order of 8000 gauss the crystalline field can be treated as a relatively small perturbation of the Zeeman effect of the free spin $S=3/2$ of the V^{++} ion. In Fig. 1 we show schematically the behavior of the energy levels as a function of magnetic field. Abragam and Pryce⁷ have shown that the Hamiltonian suitable to paramagnetic resonance experiments can be written in the form,

$$\mathcal{H} = g\beta\mathbf{H} \cdot \mathbf{S} + D[S_x^2 - \frac{1}{3}S(S+1)] + E(S_x^2 - S_y^2) + AI_xS_x + B(S_xI_x + S_yI_y),$$

where x, y, z are the principal axes of the electric field. The g factor, in general a symmetric tensor, is taken to be isotropic and equal to 1.951 ± 0.002 as has been found experimentally by Bleaney *et al.*¹ It is to be noted that both A and B are proportional to the nuclear gyromagnetic ratio. Since there are two nonequivalent ions in a unit cell for the Tutton salts, an expression of this form holds for each of the two ions. The electronic

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¹ Bleaney, Ingram, and Scovil, Proc. Phys. Soc. (London) **A64**, 601 (1951).

² C. A. Hutchison and L. S. Singer, Phys. Rev. **89**, 256 (1953).

³ Kikuchi, Sirvetz, and Cohen, Phys. Rev. **88**, 142 (1952).

⁴ R. P. Penrose, Nature **163**, 992 (1949).

⁵ B. Bleaney, Physica **17**, 175 (1951).

⁶ H. E. White, *Introduction to Atomic Spectra* (McGraw-Hill Book Company, Inc., New York, 1934).

⁷ A. Abragam and M. H. L. Pryce, Proc. Roy. Soc. (London) **A205**, 135 (1951).

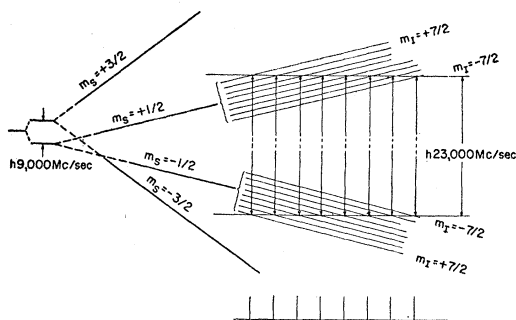


FIG. 1. Splitting of energy levels for the V^{51++} ion due to crystal-line electric field, applied magnetic field, and nuclear magnetic interaction.

part of the term values to the second approximation can be shown to be

$$\begin{aligned}
 E(-3/2 \leftrightarrow -1/2) &= g\beta H - 2(a_0 D + \frac{1}{2} b_0 E) \\
 &\quad + (24/g\beta H)(a_1 D + \frac{1}{2} b_1 E)^2, \\
 E(-1/2 \leftrightarrow 1/2) &= g\beta H - (24/g\beta H)(a_1 D + \frac{1}{2} b_1 E)^2 \\
 &\quad + (12/g\beta H)(a_2 D + \frac{1}{2} b_2 E)^2, \\
 E(1/2 \leftrightarrow 3/2) &= g\beta H + 2(a_0 D + \frac{1}{2} b_0 E) \\
 &\quad + (24/g\beta H)(a_1 D + \frac{1}{2} b_1 E)^2,
 \end{aligned}$$

where a_n and b_n are angle dependent functions defined in the Appendix. The values $D=1720$ and $E=540$ gauss have been given by Bleaney *et al.*¹ For the special case that the magnetic field is perpendicular to the tetragonal axis of one of the ions, the spectrum of this ion is given by

$$\begin{aligned}
 E(-3/2 \leftrightarrow -1/2) &= g\beta H + D + 3E \cos^2 \psi \\
 &\quad + (6/g\beta H)E^2 \cos^2 2\psi, \\
 E(-1/2 \leftrightarrow 1/2) &= g\beta H - (6/g\beta H)E^2 \cos^2 2\psi \\
 &\quad + (3/4g\beta H)(D - E \cos^2 \psi)^2, \\
 E(1/2 \leftrightarrow 3/2) &= g\beta H - D - 3E \cos^2 \psi \\
 &\quad + (6/g\beta H)E^2 \cos^2 2\psi.
 \end{aligned}$$

The effect of the nuclear magnetic moment is to split each of the above transitions into a group of $2I+1$ lines. The nuclear contributions to the term values are given, under the assumption $A=B$, by

$$\begin{aligned}
 E(-3/2, m \rightarrow -1/2, m) &= Am \\
 &\quad + (A^2/2h\nu)[I(I+1) - m^2 - 2m], \\
 E(-1/2, m \rightarrow 1/2, m) &= Am \\
 &\quad + (A^2/2h\nu)[I(I+1) - m^2], \\
 E(1/2, m \rightarrow 3/2, m) &= Am \\
 &\quad + (A^2/2h\nu)[I(I+1) - m^2 + 2m].
 \end{aligned}$$

Thus as the crystal is rotated, the center of each group of $(2I+1)$ hfs lines will shift, but the spacing of the hfs components will remain constant, within the limits of experimental observability.

2. EXPERIMENTAL PROCEDURES

The crystals of vanadium ammonium Tutton salts were prepared by a method suggested by Meyer and Aulich,⁸ and by Hutchison and Singer.² A solution composed of 10 mg of V_2O_5 , enriched to 22.83 percent of V^{50} , in 2 cc of 50 percent sulfuric acid and of 4 cc of 1 *M* ammonium sulfate was electrolyzed at ice temperature until the violet color of the divalent vanadium became pronounced. Then 4 cc of 2 *M* zinc sulfate was added and the mixture was transferred to a crystallizing vessel. All operations were carried out in a carbon dioxide atmosphere to avoid oxidation of the divalent vanadium. It was found that when crystallization took place in a slow stream of CO_2 in a vibration free room large single crystals, about 1 cm on an edge, were obtained in about 24 hours. The crystals were not analyzed for vanadium content, but in the solution the atomic V to Zn ratio was 1:80.

Crystals grown in this manner were found to have an appreciable amount of cloudy material attributed to imperfect crystallization or the occlusion of liquid. Since such material causes a large reduction in the transmitted microwave power, as well as an increase in the widths of the absorption lines, care had to be exercised so that the portion of crystal used contained only clear material. The specimens were cut from the single crystals in the form of cylinders about 2 mm in diameter and 5 mm in length, and were mounted on the end of a quartz rod extending axially into the transmission cavity. It was thus possible to rotate the crystal about the cylinder axis. The vanadium content of a single specimen is estimated at 60 micrograms, or about 7.5×10^{17} atoms.

The cylindrical cavity, 17 mm in length and in diameter, is fed by wave guides coupled to the sides of the cavity. In this way the mode TE_{011} is excited; that is, the mode in which the E field and the transverse H field vanish along the axis, while the longitudinal H field, which causes the transitions, is a maximum at the center and falls off sinusoidally to top and bottom. The frequency was held constant at 23 050 Mc/sec while the magnetic field was modulated at 60 cps with an amplitude of about 80 gauss. The transmitted power was rectified and fed into a wide-band preamplifier, the output of which was then displayed on the oscilloscope. Since the field is modulated sinusoidally while a sawtooth sweep is applied to the oscilloscope, a double pattern is observed.

The magnetic field was not measured directly. Instead a measurement was made of the magnet current necessary to display each line at the same point of the observed pattern. The current determined by the use of a standard 50-mv shunt and a Leeds and Northrup type "K-2" potentiometer, was accurate to a few ma in 15 amp. The extent of the hyperfine structure pat-

⁸ J. Meyer and M. Aulich, *Z. anorg. u. allgem. Chem.* **194**, 278 (1930).

terns for a given electronic transition was of the order of 2 amperes. To convert current readings to field values we could assume over the range of interest a relation of the form,

$$H = H_0 + ai + bi^2.$$

For the difference of the resonant fields of the hfs components $\pm m$, we have

$$2mA = g\beta(H_{-m} - H_m) = a(i_{-m} - i_m) + b(i_{-m}^2 - i_m^2).$$

By taking a number of such pairs it is possible to obtain reliable values of a and b . In this way we get the fields in units of A , the electron nuclear coupling constant. This is sufficient for comparing the experimental and theoretical relative spacings. Actual field values are obtained by taking the value of 96 gauss for A as given by Bleaney *et al.*¹

Because of the way the crystal specimens were prepared and mounted, the directions of the crystalline axes were not known precisely. In general, then, a quite complex spectrum is obtained, consisting of 48 lines for V^{51} due to the 3 electronic transitions, the 8 hfs levels, and the 2 nonequivalent ions. When the presence of a relatively small concentration of V^{50} is taken into account, with its higher nuclear-spin multiplicity and small hfs splitting, it becomes very important to strive for simplifications of the spectrum, as well as for absorptions as intense and as narrow as possible. The requirement of narrow lines practically limits one to the use of the electronic transition $-1/2 \rightarrow 1/2$; the other electronic transitions produce lines so broad that an unambiguous nuclear spin determination for V^{50} would be impossible. Some measurements were made at liquid nitrogen temperature. This produces no appreciable narrowing in this case but increases the intensity of the absorptions considerably. Satisfactory results can, however, be obtained easily at room temperature where the bulk of our measurements were made, if care is taken to find an advantageous crystal orientation. Lines were observed to have a full width at half-maximum of about 13 gauss. This is approximately that expected from the proton magnetic moments in the water of hydration and indicates that a lower V:Zn ratio would not have produced further narrowing.

A good orientation of the crystal satisfies two conditions: (1) the spectra of the two ions in unit cell coincide; (2) the hfs patterns due to different electronic transitions do not overlap. The gain in intensity by a factor of 2 and the simultaneous simplification of the spectrum which condition (1) makes possible, correspond to an orientation which can always be achieved, as can be seen with the aid of Fig. 2. The elucidation of the relations among the crystallographic, crystalline electric field, and susceptibility axis systems is due to Bleaney *et al.*¹ In Fig. 2 the z axis is the cylinder axis and also the direction of the rf magnetic field; T_1 and T_2 , the tetragonal axes of the two nonequivalent ions;

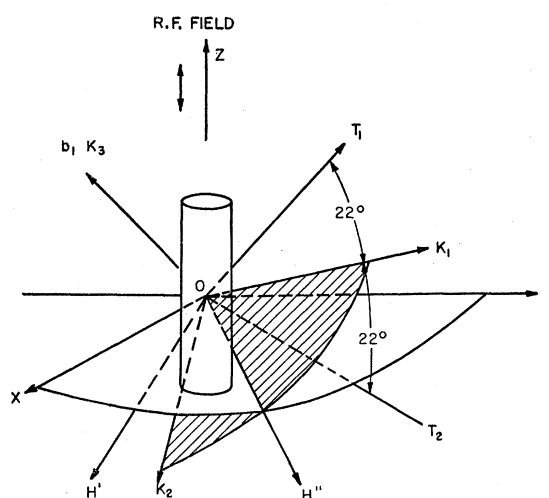


FIG. 2. Diagram showing orientations of crystalline and magnetic axes relative to the steady and rf magnetic fields.

K_1, K_2, K_3 the principal susceptibility axes; and b , coinciding with K_3 , a crystalline principal axis supposed oriented in an arbitrary way with respect to z . The plane ac (or K_1K_2) is a plane of symmetry of the crystal, a reflection in this plane carrying the one ion in unit cell into the other. Then when the magnetic field lies in this plane the spectra of the two ions coincide, since their tetragonal axes make equal angles with the field direction. Now the experimental arrangement is such that the direction of the field may be varied in the xy plane. Thus, when the field is along an arbitrary direction in this plane, say H' , the spectra of the two ions are in general separate; but when it lies along H'' , the intersection of the xy and ac planes, the two spectra coincide. This argument applies strictly only if the rhombic field vanishes. Though a small rhombic field must be assumed to analyze the spectra completely, it was found in every case that it was possible by rotating the crystal to make the two sets of $-1/2 \leftrightarrow 1/2$ lines coincide. The fact that the rhombic field is not truly zero, however, can be shown by the fact that for this particular orientation the sets of lines due to the other electronic transitions did not in general quite coincide.

To resolve the various electronic transitions, several specimens were examined. Best results were obtained with one for which the cylinder axis lay nearly along the b axis. For the orientation which gave sharpest hfs lines, the nearest unwanted line was the component at highest field of the electronic transition $-3/2 \rightarrow -1/2$ which nearly coincided with the lowest field component of $-1/2 \rightarrow 1/2$. This did not interfere with the investigation of the V^{50} hfs.

3. ANALYSIS OF THE SPECTRUM

The observed and the calculated patterns for the $(-1/2 \rightarrow 1/2)$ electronic transition are presented in

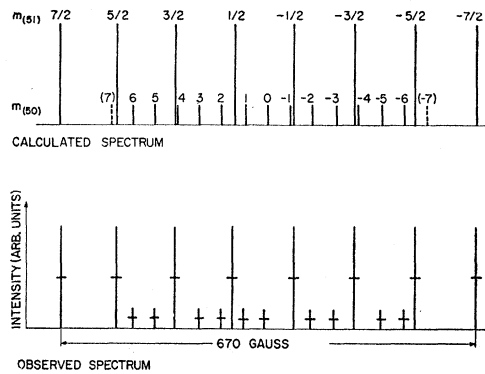


FIG. 3. The calculated and observed hfs spectrum of the electronic transition $-\frac{3}{2} \rightarrow \frac{3}{2}$ of V^{51} and V^{50} (22.83 percent abundant) for $I(V^{50})=6$ and 7. The calculated spectrum includes the second-order terms.

Fig. 3. The expected spectrum was calculated from the known spin of V^{51} , the spin 6 for V^{50} , the abundance of V^{50} , and the known ratio $g(V^{50})/g(V^{51})$. The observed spectrum consists of 8 strong lines, clearly due to V^{51} , and a number of weaker components. The latter are nearly symmetrically spaced with respect to the center of the hfs pattern, except for a small shift to higher field. From the isotopic abundances the intensity ratio of V^{50} to V^{51} lines is calculated to be 1:5.5 for $I(V^{50})=6$ and 1:6.3 for $I(V^{50})=7$. The measured ratio for the weak to strong lines was about 1:5, but the inaccuracy was such that this could not be taken as a definite indication of the spin. Further evidence that these weak lines were the hfs pattern for V^{50} came from a rough measurement of the splitting between successive components. For the weak components this is about 0.39 times the splitting for the strong lines, while the ratio of the nuclear g factors is 0.379074 ± 0.000017 .⁹ In all specimens investigated the components $m(V^{50}) = \pm 6$ were observed, together with all components of lower m except $m = \pm 4$. The latter according to theory should be unresolved from the strong lines $m(V^{51}) = \pm 3/2$. We could then immediately take $I=6$ as a lower limit for the spin of V^{50} . Figure 4 shows an oscilloscope trace of the $m = -3/2$ component for V^{51} and $m = -2$ and -3 for V^{50} .

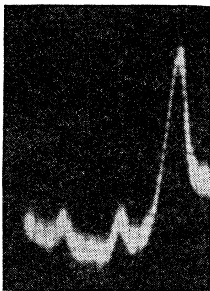


FIG. 4. Oscilloscope trace showing the components $m = -\frac{3}{2}$ for V^{51} and $m = -2$ and -3 for V^{50} .

Since there was a certain initial expectation that the spin might be 7, considerable effort was directed toward searching for the corresponding components. No line attributable to $m = \pm 7$ or to a higher m value was found. The following argument can be given to show that if these lines existed they would have been observed. Figure 5 shows the V^{51} lines $m=1/2$ and $m=-5/2$. For $m=1/2$ the satellites $m(V^{50})=2$ and 1 are separated from the main peak by 19 and 18 gauss, respectively. According to theory, these separations should be 19 and 17 gauss for a spin of 6, and 20 and 16 gauss for a spin of 7. The line $m=-5/2$ shows a single satellite with a separation of 21 gauss. This corresponds precisely to expectation for $I=6$, whereas if I were 7 this splitting would be 18 gauss, and another satellite would be present on the other side of the strong peak and separated from it by 19 gauss. Since the above discussion shows that a satellite separation of 19 gauss should be adequate for resolution if the satellite were present, we conclude that the spin of V^{50} is 6. Baker and Bleaney¹⁰ have obtained this result independently by using a similar method but a different compound of vanadium.

As has been mentioned in Sec. 2, the field units are essentially arbitrary in our method of calibration so that the calculated spectrum (Fig. 3) is plotted on this scale to fit at the lines $m(V^{51}) = \pm 5/2$. Neglecting second-order effects, there remains then only one parameter to choose to calculate the V^{50} spectrum on the same scale. This is the ratio of the nuclear-electronic coupling constants A , or equivalently the ratio of the g factors. The value that we obtain is

$$g(V^{50})/g(V^{51}) = 0.3792 \pm 0.0008.$$

This compares well with the nuclear magnetic resonance value, 0.379074 ± 0.000017 , given by Walchi and Morgan.⁹ Relatively high precision can be obtained in the measurement of g -factor ratios by paramagnetic resonance by taking symmetrical pairs of hfs components as widely spaced as possible. In our case convenient pairs were $\pm 5/2$ for V^{51} and ± 6 for V^{50} . In this way the second-order effects cancel and the effect of errors in field measurement is reduced.

The calculated spectrum includes the effect of the second-order terms. Though the effects are small they are noticeable, as can be seen by considering the separations of the V^{50} components ± 6 from the V^{51} components $\pm 5/2$. Only a rough value of the coupling constant B can be obtained in this way. Within experimental error it is equal to A , so that as Bleaney *et al.*¹ report the hfs coupling is isotropic.

4. DISCUSSION

The spin $I=6$ determined for V^{50} is just one less than the maximum value allowed by shell theory. The nu-

⁹ H. E. Walchi and H. W. Morgan, Phys. Rev. **87**, 541 (1952).

¹⁰ J. M. Baker and B. Bleaney, Proc. Phys. Soc. (London) **A65**, 952 (1952).

cleus has 23 protons and 27 neutrons, both odd particles being in the state $f_{7/2}$. According to Nordheim¹¹ the spin of an odd-odd nucleus is given by $|j_1 - j_2| < I < |j_1 + j_2|$, where j_1 and j_2 are the spins of the odd nucleons. Furthermore, a value close to the maximum is favored if the odd nucleons belong to the same Schmidt line. Thus for V^{50} a spin of 7 was expected, though the smaller value obtained does not represent an exception to a rigorously valid rule. For Cl^{36} and K^{40} , a spin one less than the maximum allowed value has been found, whereas the spins of lighter odd-odd nuclei, with the exception of Li^6 , have the maximum value allowed by shell theory. Recently Hitchcock¹² has reported that calculations for V^{50} using a δ -function potential give the state with $I=6$ as the lowest level. The gyromagnetic ratio of V^{50} agrees remarkably well with that predicted by shell theory. If two angular momentum vectors, j_1 and j_2 , contribute to the magnetic properties of a nucleus, the gyromagnetic ratio of the nucleus is given by

$$g = \frac{1}{2} \left\{ g_1 + g_2 + (g_1 - g_2) \frac{[j_1(j_1+1) - j_2(j_2+1)]}{I(I+1)} \right\}.$$

Applied to the $f_{7/2}$ proton, with $j_1 = L = 3$, $g_1 = 1$, $j_2 = 1/2$, $g_2 = g_p$, we obtain $g(f_{7/2}, p) = 1.655$. For the $f_{7/2}$ neutron we have $j_1 = L = 3$, $g_1 = 0$, $j_2 = 1/2$, $g_2 = g_n$, giving $g(f_{7/2}, n) = -0.547$. For the V^{50} nucleus then, with $j_1 = j_2 = 7/2$, $g_1 = 1.655$, $g_2 = -0.547$, we obtain $g(V^{50}) = 0.554$. This good agreement, in spite of the uncertainty in the effective magnetic moments of the proton and neutron bound in the nucleus, stems from the fact that the correction to their magnetic moments cancel for odd-odd nuclei consisting of proton and neutron shells of the same spin and parity. This has been pointed out by Talmi.¹³

We wish to express our appreciation to the Stable Isotope Production Division of the Atomic Energy Commission for making the enriched vanadium sample available to us; to Dr. Simon Freed and Dr. Norman Elliott of this Laboratory and Dr. L. Singer of the Naval Research Laboratory for many helpful discussions on the chemistry of vanadium; and to Mr. John Ciperano for technical assistance. One of us (C.K.) is grateful for the leave of absence from Michigan State College, during which this investigation was carried out.

¹¹ L. W. Nordheim, *Revs. Modern Phys.* **23**, 322 (1951).

¹² A. Hitchcock, *Phys. Rev.* **87**, 664 (1952).

¹³ I. Talmi, *Phys. Rev.* **83**, 1248 (1951); see A. deShalit, *Helv. Phys. Acta* **24**, 296 (1951).

APPENDIX

To calculate the term values it is convenient to take the axis of quantization along the steady magnetic field H and transform the operators defined in the crystalline principal axis system xyz to the system $x'y'z'$. This is readily done by taking the representations,

$$S_+ = \xi \partial / \partial \eta, \quad S_z = \frac{1}{2} (\xi \partial / \partial \xi - \eta \partial / \partial \eta), \quad S_- = \eta \partial / \partial \xi,$$

where ξ and η transform as

$$\xi = \alpha \xi' - \beta^* \eta', \quad \eta = \beta \xi' + \alpha^* \eta',$$

with

$$\alpha = \cos(\theta/2) e^{i(\phi+\psi)/2}, \quad \beta = i \sin(\theta/2) e^{i(\phi-\psi)/2}.$$

Then

$$\begin{aligned} S_+ &= \xi \partial / \partial \eta = \alpha^2 \xi' \partial / \partial \eta' - \alpha \beta^* (\xi' \partial / \partial \xi' - \eta' \partial / \partial \eta') \\ &\quad - \beta^{*2} \eta' \partial / \partial \xi' \\ &= \alpha^2 S_+' - 2\alpha \beta^* S_z' - \beta^{*2} S_-'. \end{aligned}$$

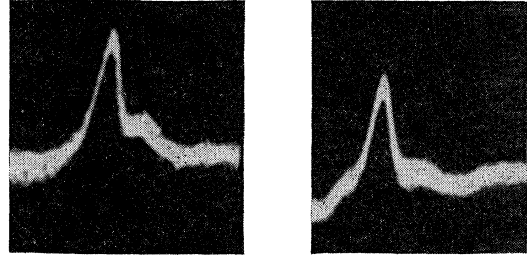


FIG. 5. Photographs of two sections of spectrum. (a) The line $m = -5/2$ of V^{51} and the satellite $m = -6$ of V^{50} ; (b) lines $m = 1/2$ of V^{51} and $m = 1$ and 2 of V^{50} .

The transformation properties of S_z' and S_-' can be obtained in a similar manner. The tetragonal and rhombic field terms then become

$$\begin{aligned} D[S_z'^2 - \frac{1}{3} S(S+1)] \\ = D[a_2 S_+'^2 + a_1 (S' S_+' + S_+' S') + a_0 \{ S_z'^2 - \frac{1}{3} S(S+1) \} \\ + a_{-1} (S' S_-' + S_-' S') + a_{-2} S_-'^2], \end{aligned}$$

$$\begin{aligned} E(S_z'^2 - S_y'^2) = \frac{1}{2} E(S_+'^2 - S_-'^2) = \frac{1}{2} E[b_2 S_+'^2 \\ + b_1 (S_z' S_+' + S_+' S_z') + b_0 \{ S_z'^2 - \frac{1}{3} S(S+1) \} \\ + B_{-1} (S_z' S_-' + S_-' S_z') + b_{-2} S_-'^2], \end{aligned}$$

where,

$$a_2 = a_{-2}^* = -\frac{1}{4} \sin^2 \theta e^{-2i\phi},$$

$$a_1 = a_{-1}^* = \frac{1}{2} i \sin \theta \cos \theta e^{-i\phi},$$

$$a_0 = \frac{1}{2} (3 \cos^2 \theta - 1),$$

$$b_2 = b_{-2}^* = [\cos^4(\theta/2) e^{-2i\psi} + \sin^4(\theta/2) e^{2i\psi}] e^{-2i\phi},$$

$$b_1 = b_{-1}^* = i \sin \theta [\cos^2(\theta/2) e^{-2i\psi} - \sin^2(\theta/2) e^{2i\psi}] e^{-i\phi},$$

$$B_0 = -3 \sin^2 \theta \cos(2\phi).$$

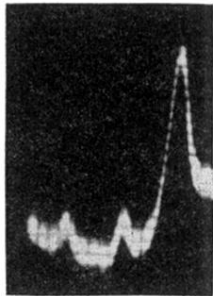
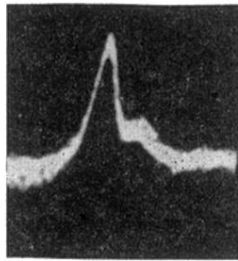
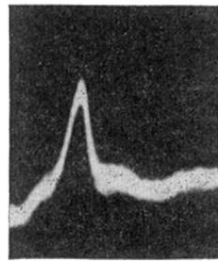


FIG. 4. Oscilloscope trace showing the components $m = -\frac{3}{2}$ for V^{61} and $m = -2$ and -3 for V^{60} .



(a)



(b)

FIG. 5. Photographs of two sections of spectrum. (a) The line $m = -5/2$ of V^{61} and the satellite $m = -6$ of V^{60} ; (b) lines $m = \frac{1}{2}$ of V^{61} and $m = 1$ and 2 of V^{60} .