Hyperfine Structure of the Spectral Lines and Nuclear Spins of U^{233} and Pu^{239}

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The hyperfine structure in 12 lines of the U^{233} was investigated. In four cases the maximum total of six components was observed. It was determined that the spin $I_{233} = 5/2$. A deviation from the interval rule was detected, which is explained by the presence of a nuclear quadrupole moment in U^{233}.

Approximately 70 lines with doublet hyperfine structure were discovered in the Pu^{239} spectra. The spin $I_{239} = 1/2$. The width of the hyperfine splitting varied from 0.04 to 0.2 cm^{-1}. In certain spectral lines the stronger component was located on the long wavelength side and in other lines, on the contrary, on the short wavelength side. For principal sublevels this corresponds to a transition from an upper non-split level to a lower split level, and conversely.

1. INTRODUCTION

THE study of the hyperfine structure of the spectral lines of the actinide elements presents great interest. It is known that in many cases the hyperfine structure of the lines suffices provide a reliable determination of the nuclear moments, which are requisites for further development of nuclear shell structure, theory of isomers and beta disintegration, and a series of other questions of nuclear physics. It ought to be noted, however, that up to now researches on the hyperfine structure and nuclear moments of isotopes of the actinide elements are far from complete. According to publications in foreign literature, the nuclear spins of Pa^{231}[1], U^{235}[2], Np^{237}[3], and Am^{241}[4] were determined at the beginning of the present year. In addition, at the end of 1952 and the beginning of 1953 the nuclear spins of U^{233} and Pu^{239} were measured by us and, independently, by S. E. Frisch, N. I. Kalitievskii, and M. P. Chaika. Later in the year our results were confirmed by references 5,6. In reference 5 data are also published concerning the magnetic quadrupole moments of U^{235}. In the present paper results are reported on the determination of the spins of U^{233} and Pu^{239} and, also on our investigations of the hyperfine structure of the lines of the mentioned isotopes.

[1] H. Schuler and H. Gollnow, Naturwiss 22, 511 (1934)

[2] O. E. Anderson and H. E. White, Phys. Rev. 71, 911 (1947); G. L. Stukenbrocker and J. R. McNally, Opt. Soc. Am. 40, 336 (1950)

[3] F. S. Tomkins, Phys. Rev. 73, 1214 (1948)

[4] M. Fred and F. S. Tomkins, Phys. Rev. 89, 318 (1953)

[5] K. L. V. Sluis and J. R. McNally, Opt. Soc. Am. 44, 87 (1954)

[6] M. van den Berg, P. F. A. Klinkenberg and P. Regnaut, Physica 20, 37 (1954)

2. EXPERIMENTAL PART

A discharge tube with a hollow cathode was utilized for the excitation of the spectra of uranium and plutonium. A schematic representation of this tube is given in Fig. 1. The essential part of the tube is the all-metal aluminum cylinder *M*, having in the bottom part a cylindrical cavity *K*, of 5 mm diameter and 10 mm depth, in which the experimental sample is placed. The cylinder *M* is the cathode, in the cavity of which the electric discharge is concentrated. The anode *A* is also made of aluminum. It is a small cylindrical cup with a hole in the bottom for the passage of a light beam. Such construction of the anode permits a more complete recovery of the sample, since, in the main, the sample settles out on the bottom. The anode is connected through the outlet *L* to a high voltage rectifier. The glass tube *N* is incased within cylinder *M* and is bonded to it by the use of picein. This tube *N* has two branches, *F* and *E* (pure gas enters at the opening *F*, impure gas leaves the tube through opening *E*). A circulating inert gas on one side promotes the excitation of luminescence of the test sample and on the other side (at the right time), cleans the discharge tube of contamination. For cooling, the discharge tube was placed in a vessel of water. Such a light source gives sufficiently sharp and intense lines and permits one to work with a small amount of material.

The discharge tube was connected to the vacuum system in the manner shown in Fig. 2. A high vacuum (10^{-4}-10^{-5} mm Hg) was obtained by use of mercury vapor pump *I* in steps. The circulation of

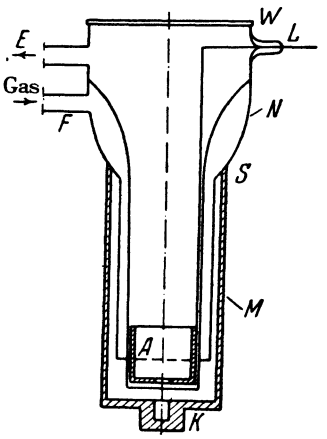

Fig. 1. Discharge Tube

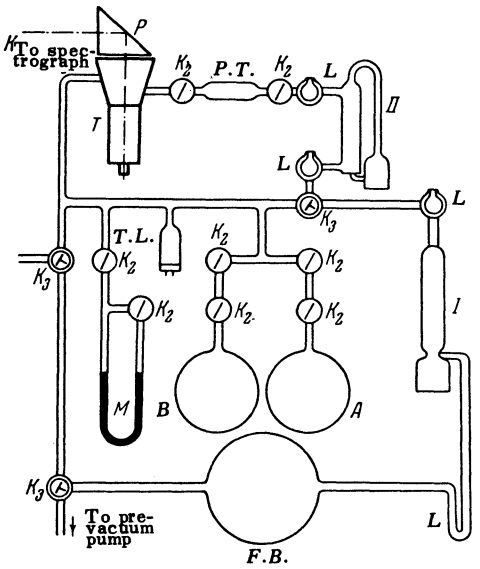

Fig. 2. Scheme of the vacuum arrangement. *T*-discharge tube, *I*-mercury vapor pump, *II*-circulating mercury vapor pump, *P.T.*- potassium trap, *L*-trap for mercury, *T.L.*- thermocouple lamp, *M*-oil manometer, *A* and *B*-balloons filled with gas, *F.B.*-pre-vacuum balloon, K_2-two way stopcock, K_3- three way stopcock, *P*-prism of complete internal reflection

an inert gas through the tube was accomplished by using another mercury vapor pump, *II*. The construction of the circulating pump is such that it can function in pressures of a few millimeters of mercury. The circulating current of such inert gases as argon and krypton was cleaned in a potassium trap. The potassium trap was a glass balloon, on the internal surface of which was applied a fine layer of metallic potassium. For work with helium it is necessary to provide a carbon trap. For measuring the pressure in the discharge tube in the interval from 0.1 to 5 x 10^{-4} mm Hg, a thermocouple manometer (UTV-49 with lamp LT-2) was used. An oil manometer was used for measuring higher pressures.

In our experiments a Fabry-Perot interferometer (IZS-9), prepared in GOI, with half-silvered glass plates was employed as the high resolving power instrument. For preliminary dispersion, a three prism glass Shteinkhel spectrograph was used with a camera having an objective of focal length F = 640 mm. The Fabry-Perot interferometer was placed between the light source and the slit of the spectrograph. The interference pattern was projected onto the widened slit of the spectrograph by using a high quality Tessar objective (F = 600 mm).

Pure isotopes in the combined oxide forms, U_3O_8 and PuO_2, were used for the investigations of the hyperfine structure of the spectral lines of U^{233} and Pu^{239}. Samples of weight from 3.5 to 8 mg were inserted into the discharge tube. It appeared that uranous oxide and the plutonium oxide in the discharge tube were not immediately excited, even though different inert gases, He, A, Kr, and mixtures of them, were used for the final filling. In order to excite the spectra of uranium and plutonium, we had to reduce initially the uranous oxide and the plutonium oxide to the metallic forms. This reduction was carried out in the discharge tube with atomic hydrogen, under conditions of an electric discharge with current strength near 0.3 amp in the presence of a hydrogen pressure of the order of 2 mm Hg. A time of 1-5 hours was required for reduction to the metal. The tube was fed with a constant current from a high voltage rectifier. Optimum conditions for the excitation of uranium and plutonium are: argon pressure, 2 mm Hg; current strength, 0.2 amp. Hyperfine structure in the spectra of uranium was photographed with interferometer plate separations of 6,10 and 15 mm, with exposures of 20 min. Hyperfine structure in the spectra of plutonium was obtained with separation of 10 and 15 mm and exposures of 30 min.

Hyperfine structure of the lines of uranium and plutonium was measured with an ABBE comparator Central rings located on one side of the interference pattern were used for the measurement of the hyperfine structure of the lines of uranium. The method of Ritschel-Schrammen[7,8] was used for

[7] R. Ritschel, Z. Physik. 79, 1 (1932)

[8] S. Tolansky, *High Resolution Spectroscopy*, London, 1947. Ch. 9

finding the diameters of the circles in this case. Treatment of the measured results proceeded by the method of "right-angled tables" [8] Non-central rings were utilized for measurement of the hyperfine structure of the lines of plutonium. The approximate method of Mc Nair[8] was used for treatment of the measured results.

3. RESULTS

A) HYPERFINE STRUCTURE OF THE LINES OF U^{233}

Careful analysis of the U^{233} spectrogram permitted us to observe marked hyperfine structure in 12 lines. Among them a complete resolution of the six-component hyperfine structure was found on five lines: 6826.93; 5976.34; 5915.40; 4515.28 and 4171.59 A. In the remaining lines there was observed either a partial resolution, or strong broadening of the contours of the lines. Combined data on the hyperfine structure of these lines are presented in Table I. Our measurements for the complete range of the splitting are presented in the fourth column of this table; the type of splitting is shown in the fifth column. "Complete" splitting means that six distinct components are observed in the spectrogram; "partial" splitting shows that the structure of the lines, under our conditions, is not fully obtained. The direction of the component of decreasing intensity is recorded in the sixth column. "Red" decrease means that the intensity of the components is diminished on the side of the decreasing wave numbers; "violet" decrease means that the intensity of the components decreases on the side of increasing wave numbers. It is seen from the table that the splitting in spark lines is larger than in arc lines. Among the arc lines the largest width of splitting (0.32 cm^{-1}) is observed for line 5915.40 A; among the spark lines the line 4515.28 A experiences the largest splitting (nearly 0.7 cm^{-1}).

TABLE I
Hyperfine structure in the spectra of U^{233}

Wave length in A	Type of atom	Classification[9]	Total width of the splitting (cm^{-1})	Type of splitting	Direction of decreasing intensity
6826.93	I	$f_3ds^2\ {}^5L_6^0 - 146_6$	~0.15	Complete	Violet
6449.16	I	${}^5K_5^0 - 161_4$	~0.18	Partial	"
6077.29	I	${}^5K_5^0 - 170_6$	—	"	—
5976.34	I	$A_7^0 - 205_8$	~0.25	Complete	Red
5915.40	I	${}^5L_6^0 - 169_7$	0.323	"	"
5564.17	I	$A_7^0 - 218_6$	~0.2	Partial	Violet
4543.63	II	$f_3ds\ \ K^9/_2 - 229^{11}/_2$	~0.5	"	Red
4515.28	II	$L^{11}/_2 - 224^9/_2$	~0.7	Complete	"
4472.33	II	$L^{11}/_2 - 226^{11}/_2$	—	Partial	"
4341.69	II	$L^{11}/_2 - 233^9/_2$	—	"	"
4171.59	II	$L^{13}/_2 - 257^{13}/_2$	~0.6	Complete	Violet
4090.13	II	$L^{13}/_2 - 261^{13}/_2$	~0.5	Partial	—

It is not difficult to make a deduction based on the six-component hyperfine structure of the four completely resolved lines concerning the value of the nuclear spin of U^{233} If it is assumed that $J < I$, the terms ought to split into $2J + 1$ sublevels. This means that each of the investigated lines of uranium should split, not into six components, but into a considerably larger, and moreover, variable, number of components. It is easy to see that arc lines should split into an odd number of components and spark lines into an even number of components. Insofar as this contradicts experiment, the assumption $J < I$ must be considered untrue. Another possibility remains, namely that $I < J$. In this case each term will split into $2I + 1$ sublevels of hyperfine structure. If one assumes that for each observed transition either the upper or the lower level experiences splitting, then the split lines ought to be composed of $2I + 1$ components. On the basis of our experiment, it follows that $2I + 1 = 6$, hence the nuclear spin of U^{233} equals 5/2.

The hyperfine structure of U^{233} in the arc line

[9] C. C. Kiess, C. J. Humphreys and D. D. Laun, J. Nat. Bureau Stand. 37, 57 (1946); J. C. Van den Bosch, Physica 15. 503 (1949)

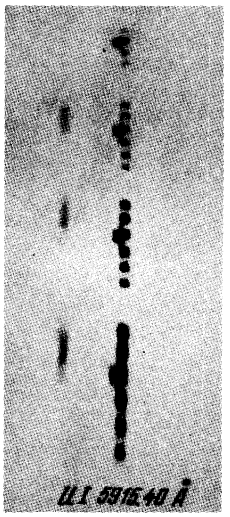

Fig. 3. Hyperfine structure of the line UI 5915.40 A

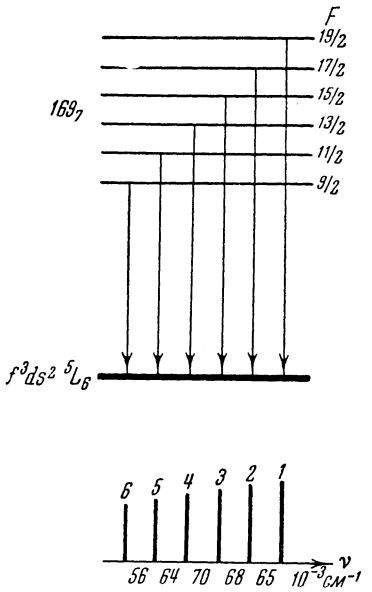

Fig. 5. Transition scheme and hyperfine structure of the line UI 5915.40 A

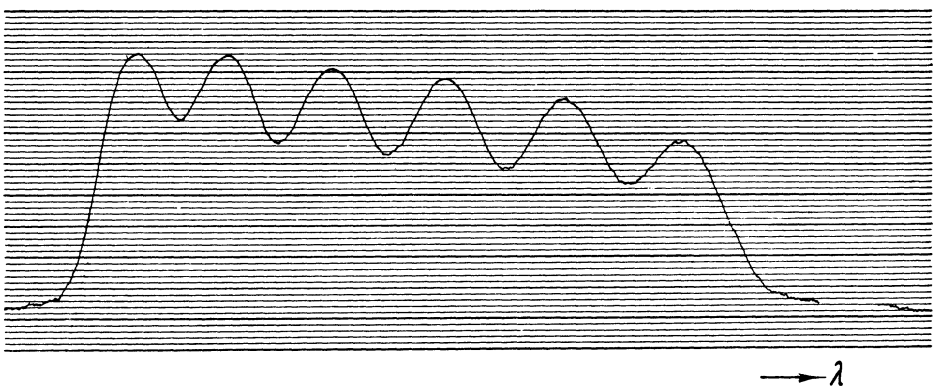

Fig. 4. Microphotograph of the line UI 5915.40 A

5915.40 A was studied by us in the most detail. In Fig. 3 there is shown a spectrogram of this line, obtained with an interval of 10 mm between plates of the interferometer. Fig. 4 shows a microphotograph of the hyperfine structure of this line, recorded by using a registering microphotometer. The six components structure, with the components gradually decreasing in intensity on the side of decreasing wave numbers, is clearly seen in the photographs. At the top of Fig. 5 there is a scheme of transitions, and below, a diagrammatic structure of the line 5915.40 A. The scheme of the transitions is based on the experimental determination of the direction of decrease of intensity of the components of the hyperfine structure, in view of the intensity rule, and on the assumption that only the upper term experiences splitting. From this scheme we see that the term 169_7 appears normal Our assumption concerning the splitting of the upper term in the case of line 5915.40 A is confirmed by the fact that line 6826.93 A, with the same lower term, has half the width of splitting and an inverted decrease of intensity in the components of the hyperfine structure.

By use of the blackening of the marks, we found a relation for the intensities of the components of the hyperfine structure for line 5915.40 A which was shown equal to : $i_1 : i_2 : i_3 : i_4 : i_5 : i_6 = 34 : 30 : 28 : 25 : 21 : 19$. Inasmuch as the times of exposure for production of the darkened marks and plotting of the structure were different, this was assumed to be a Shwartzchild constant. Moreover, the variation of intensities in the interference pattern from order to order was calculated; and a correction was made for the superposition, one on the other, of the edges of the hyperfine structure components. The distribution of intensities along the contour of the interference maxima was found, for the non-splitting argon lines, to be located near the investigated uranium lines. The discovered relation for intensity shows that within the limits of error of the measurements, the intensity rule is obeyed.

The intervals between components of the hyperfine structure, beginning with the long wavelength components, were shown equal to : 0.056; 0.064; 0.070; 0.068; 0.065 cm^{-1} (Fig. 5). The complete width of the structure comprises 0.323 cm^{-1}. If one proceeds from the splitting of the upper term, then according to the interval rule, the relation for the spacing between components ought to be equal to: 56: 66.2 : 76.4; 86.5: 96.7. From a comparison of the experimental and theoretical data it is seen that the discovered intervals between components are not contained in the interval rule. This can be explained by the presence of a quadrupole moment in the U^{233} nucleus. If one neglects the excitations of lower terms of the hyperfine structure, the discovered intervals for the line 5915.40 A are contained in the Eq.[10]:

$$\Delta T = \frac{A}{2} C + B \frac{{}^3/_8 C(C+1) + {}^1/_2 IJ(J+1)(I+1)}{IJ(2I-1)(2J-1)}$$

where $A = +0.009$ and $B = -3.8 \times 10^{-2}$. In this formula ΔT is the displacement of the lower term, expressed in cm^{-1} ; $C = F(F+1) - I(I+1) - J(J+1)$; A is a constant, characteristic of the magnetic interaction of the nucleus with the electron shells; B is a constant electric quadrupole interaction.

Comparing data from reference 2 on the hyperfine structure of line 5915.40 A for U^{235} with our data for U^{233}, it is possible to draw the conclusion that the magnetic moments of these isotopes have opposite signs. According to reference 5 the magnetic moment of the U^{235} nucleus has a negative sign; consequently, for the magnetic moment of U^{233} it is necessary to write a positive sign. Judging from the overall width of splitting of the line 5915.40 A for U^{233} and U^{235} [2], it is possible to show that the magnetic moment of U^{233} is approximately 1.5 times larger than the magnetic moment of U^{235}

B) Hyperfine Structure of the Lines of Pu^{239}

Careful analysis of the spectrogram obtained for the study of the hyperfine structure of plutonium showed that approximately 70 lines are split into two components. Of these the following lines were the clearest: 4196.16; 4206.37; 4289.10; 4393.87; 4396.31; 4406.73; 4441.57; 4456.61; 4468.48; 4472.70; 4504.80; 4535.95; 4630.82; 4664.10 A. As an example we reproduce (Fig. 6) one of the sections of plutonium spectra with lines possessing the characteristic doublet hyperfine structure.

Proceeding from the doublet hyperfine structure of the spectral lines of plutonium, it is easy to determine the mechanical nuclear moment. Let us assume that $J < I$; then the terms ought to split into $2J + 1$ sublevels. If one assumes that for each observed transition either the upper or the lower level experiences splitting, the lines ought to be split into $2J + 1$ components. Inasmuch as both arc and spark lines, having different integral and half integral values J, are present in the spectra of plutonium, the different lines ought to split into different numbers of components (arc lines into an odd number of components; spark lines into an even number of components). In our case, however, all split lines have only two components. Consequently, the assumption $J < 1$ falls away and there remains the other possibility, that $J \geq I$. In this case each term will split into $2I + 1$ sublevels of hyperfine structure. Hence, the mechanical nuclear moment of Pu^{239} equals 1/2.

We measured the intervals between components for all lines with this structure. Results of the measurements are presented in Table 2. It was shown that the overall width of the hyperfine structure ranges from 0.04 to 0.2 cm^{-1}. The greatest splitting is found in the lines 4097.51; 4468.48; 4472.70; 4504.80 A, and the least in lines 4753.47; 5549.49; 5562.06; 6091.92; 6119.25; 6488.84 A. Under the conditions of our experiment approximately one hundred lines of plutonium do not give hyperfine splitting. Of these the most intense are: 4101.91; 4167.57; 4189.97; 4335.91; 4385.37; 4419.34; 4627.39; 4689.40; 4731.02 A.

The majority of the split lines of plutonium have components with nearly similar intensities. However, the order of the component lines of the

[10] H. Casimir, Physica 2, 719 (1935)

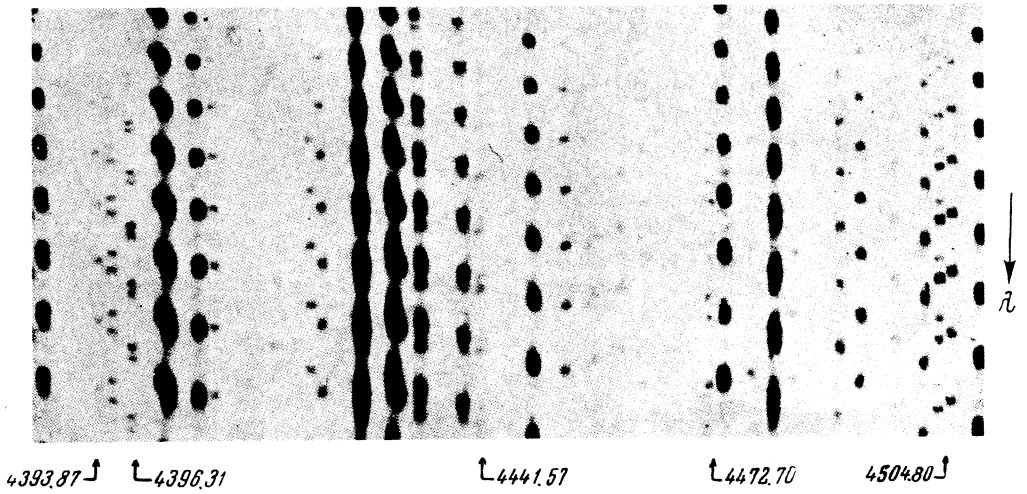

Fig. 6. Hyperfine structure of the lines of plutonium

TABLE II

Lines of plutonium possessing hyperfine structure

Wave length in A	Intensity in Arc	Width of hyperfine structure		Wave length in A	Intensity in Arc	Width of hyperfine structure	
		in cm^{-1}	in A			in cm^{-1}	in A
4064.65	4	0.098	0.016	4691.96	2	0.154	0.034
4097.51	4	0.198	0.033	4698.63		0.101	0.022
4141.22	4	0.158	0.027	4701.04	5	0.121	0.027
4196.16	7	0.111	0.019	4715.64	2	0.08	0.018
4206.37	7	0.128	0.023	4720.62	2	0.06	0.013
4211.96	3	0.119	0.021	4735.31	2	0.097	0.022
4224.02	2	0.116	0.021	4741.82	1	0.089	0.020
4229.63	5	0.130	0.023	4753.47	1	0.048	0.011
4249.63	3	0.118	0.021	4808.89	3	0.116	0.027
4269.64	2	0.073	0.013	4810.54	3	0.061	0.014
4289.10	2	0.115	0.021	4823.04	7	0.133	0.031
4316.61	3	0.143	0.027	4842.40		0.167	0.039
4326.23	2	0.136	0.025	4957.4		0.161	0.040
4358.00	6	0.139	0.026	5014.15		0.136	0.034
4392.73	4	0.089	0.017	5023.28	5	0.089	0.022
4393.87	8	0.134	0.026	5270.01		0.114	0.032
4396.31	8	0.09	0.017	5328.35		0.149	0.042
4406.73	8	0.066	0.013	5341.98		0.149	0.042
4441.57	7	0.101	0.020	5358.44	1	0.074	0.021
4456.61	5	0.127	0.025	5498.42		0.081	0.024
4468.48	4	0.161	0.032	5549.49	2	0.038	0.012
4472.70	10	0.167	0.033	5562.06	4	0.052	0.016
4491.61	3	0.091	0.018	5570.43	3	0.121	0.037
4496.73	2	0.154	0.031	5667.29	1	0.100	0.032
4504.80	10	0.165	0.033	5733.19		0.059	0.019
4507.63		0.078	0.016	5838.92	2	0.054	0.018
4512.75		0.094	0.019	5864.83	1	0.062	0.021
4527.57	2	0.097	0.020	6091.92	4	0.050	0.018
4606.81	4	0.236	0.050	6100.32	5	0.060	0.022
4625.62	2	0.142	0.030	6119.25	4	0.044	0.016
4630.82	4	0.163	0.035	6176.21	4	0.071	0.027
4639.28	3	0.142	0.031	6192.63	7	0.039	0.015
4664.10	6	0.139	0.030	6449.6		0.064	0.027
4676.18	1	0.092	0.020	6488.84	5	0.048	0.020
4689.40	2	0.067	0.015	6542.1		0.051	0.022

hyperfine structure is strongly dependent on the intensity. In some of these the long wavelength component is brighter, in others, on the contrary, the short wavelength component is brighter. As an example we have reproduced in Fig. 7 a microphotograph of the hyperfine structure of the lines 4206.37 and 4393.87 A in two orders, obtained by using a recording Zeiss microphotometer. Such microphotographs were utilized by us for determining the difference of blackening and comparative intensities of the components of the hyperfine structure.

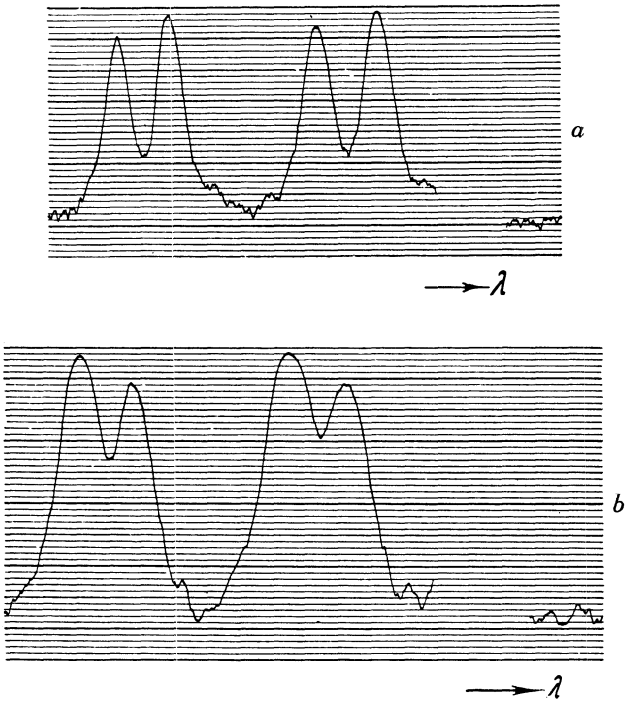

Fig. 7. Microphotographs of the lines of plutonium: $a - \lambda = 4393.87$ A; $b - \lambda = 4206.37$ A

The results of the measurements for five lines of plutonium are presented in Table 3. In the second column are given the differences of blackening between the long wavelength and short wavelength components of the hyperfine structure ($S_g - S_k$). The sign (+) means that the long wavelength component appears more intense in comparison to the short wavelength component; the sign (-) means the inverse relation of intensities. Knowing the degree of darkening of the components of the hyperfine structure, we estimated the comparative intensities of these components, I_g/I_k (see the third column of Table 3), by using characteristic curves, plotted according to the blackened marks of iron spectra, obtained by a nine-fold reduction.

TABLE III

Wave length in A	Difference of darkening $S_g - S_k$	Comparative intensity	
		I_g/I_k	I_B/I_A
4206.37	−0·33	0.49	2.04
4393.87	+0·08	1.25	1.25
4396.31	−0·14	0.75	1.33
4468.48	+0·12	1.37	1.37
4504.80	+0·29	1.87	1.87

It is necessary to explain that in the splitting of the levels (upper or lower) the long wavelength component is brighter in some cases and weaker in others. For the principal sub-levels of the hyperfine structure, the brighter long wavelength component results in transitions from an upper non-split level to a lower split level (Figure 8 A); and on the contrary, the brighter short wavelength component corresponds to transitions from an upper split level to a lower non-split one (Figure 8 b).

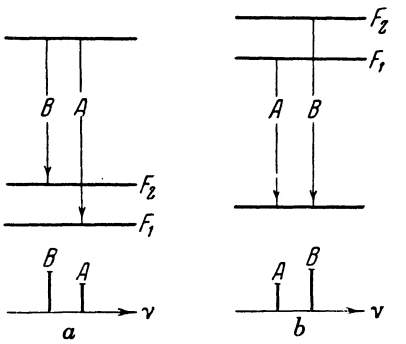

Fig. 8

In the first case $I_g/I_k > 1$; in the second, on the contrary, $I_g/I_k < 1$.

If the hyperfine structure of the lines is determined by the splitting of only one level, the intensities of the hyperfine components will be proportional to the statistical weights of the hyperfine sub-levels. In this case one can write (Fig. 8)

$$\frac{I_B}{I_A} = \frac{2F_2 + 1}{2F_1 + 1}.$$

Since for the plutonium nucleus, $I = 1/2$, then $F_1 = J - 1/2$, $F_2 = J + 1/2$. Hence we obtain :

$$\frac{I_B}{I_A} = \frac{J+1}{J}.$$

In this fashion, knowing the exact relation for the

intensities of two components of the hyperfine structure, one can find the numerical value of the quantum number J for split levels, and on the basis of this number determine the quantum numbers F_1, F_2, characterizing the total angular momentum of the whole atom. From the derived formulas it follows that if $I_B/I_A \approx 1$ (approximately equal intensity components), then the split level of the given lines will be characterized by a large value of the quantum number J; and, on the contrary, if $I_B/I_A > 1$ and attains a value in the interval $(1.5 - 3.0)$ (component intensities strongly differing), then the split level will not have a large value for the size of the quantity J.

It follows from our data that if the terms are principal ones in the case of transitions corresponding to the lines 4206.37 and 4396.31 A, there is a split upper term; and on the contrary, in the case of lines 4393.87; 4468.48; 4504.80 A, there is a split lower term. It is possible, apparently, to consider that for the upper term of the line 4206.37 A and the lower term of the line 4504.80 A. the quantum numbers are $J = 1$, $F_1 = 1/2$ and $F_2 = 3/2$. It ought, nevertheless, to be noted that insofar as the majority of the split lines of plutonium have components of similar intensity, a large part of the terms are characterized by large values for the size of the numbers J.

Very simple deductions show, that for doublet structure of the levels, transitions between split levels in some cases can give three and four-component structure lines. Carefully scanning our spectrograms, we found only one line with four components (4521.04 A) and one with three (4535.95 A). Nevertheless, the structure of these lines ought to be investigated more thoroughly, since it is necessary to keep in mind that it is possible in each of these cases to have a superposition of two different lines, close in wavelength.

If one proceeds from the size of the spin of Pu^{239}, that has been determined, then, in correspondence with theory, the quadrupole moment of this nucleus equals zero.

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83

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The Problem of the Effect of Concentration on the Luminescence of Solutions

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Improved experimental data are obtained on the effect of concentration on the luminescence of solutions of fluorescent dyes, in connection with their optical properties. It is shown that the theory of resonance excitation energy, correlating the transfer probability with the optical properties of molecular interactions, permits an approximate calculation of the depolarization constants which were previously introduced as empirical data, and also shows the relation between extinction and decrease of the duration of fluorescence.

I. INTRODUCTION

THE influence of the concentration of solutions on fluorescence is already noticeable at relatively small concentrations as concentration depolarization of fluorescence, concentration quenching and decrease of the duration of fluorescence, and is being explained as a resonance transfer of excitation energy between the molecules of the solutes. Vavilov[1-6] developed the general semiphenomenological theory of these effects, in which

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