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THE ABSORPTION SPECTRUM OF PuF_3

Ralph D. McLaughlin
(Thesis)

December, 1953

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THE ABSORPTION SPECTRUM OF PuF₃*

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December, 1953

ABSTRACT

The absorption spectrum of PuF₃ was measured over the region 2500 to 9000 Angstroms. Six constant differences were found of less than 1000 cm⁻¹. It was not possible to determine if any of these were due to low lying levels.

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

THE ABSORPTION SPECTRUM OF PuF_3

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December, 1953

I. INTRODUCTION

Magnetic evidence strongly suggests that the ground terms of Pu^{+3} and Am^{+3} are ${}^6\text{H}_{5/2}$ and ${}^7\text{F}_0$ respectively, analogous to Sm^{+3} and Eu^{+3} . This evidence is as follows.

The magnetic susceptibility of PuF_3 measured by Elliott and Lewis¹ from 76° - 300° K and extended to 600° K by Dawson, Mandlberg and Davies² agrees fairly well with the equation

$$\chi_m = \frac{0.1241}{yT(3 + 4e^{-7y} + 5e^{-16y} + \dots)} [2.14y + 3.67 + (42.9y + 0.82)e^{-7y} + (142y - 0.33)e^{-16y} + \dots]^*, \quad (1)$$

the theoretical relation deduced by Van Vleck³ for the "intermediate" case of Sm^{+3} , where the J multiplet intervals are comparable to KT . For Pu^{+3} $y = 267/T$. Over the temperature range of experimental observation the population of the lower states shifts according to the Boltzmann distribution relation and the multiplicities of the states involved.

Similarly, in work which has not yet been published in the open literature, Crane and Cunningham found that the temperature dependence of AmF_3 obeyed a relation of the form

*Corrected to current value of 6.023×10^{23} for Avagadro's number.

$$\chi_m = \frac{0.1241}{xT(1 + 3e^{-x} + 5e^{-3x} + 7e^{-6x} + \dots)} [24 + (13.5x - 1.5)e^{-x} + (67.5x - 2.5)e^{-3x} + (189x - 3.5)e^{-6x} + \dots] \quad (2)$$

as deduced by Van Vleck³ for Eu^{+3} . For Am^{+3} $x = 570/T$.

Relations (1) and (2) above may be derived in a straightforward fashion on the assumption that the susceptibilities arise from unperturbed electronic motions of electrons in "f" orbitals, with Russell-Saunders coupling between spin and orbital momenta and with the energetic order of states obeying Hund's rules.

Actual energy differences between the levels of a multiplet may be taken directly from spectroscopic data, but such data are not available for the ions in question. The multiplet intervals for the rare earths are based on effective nuclear charge for these ions, estimated from the frequencies of x-ray emission lines for heavier elements with completed "4f" shells. The intervals for the heavy elements have been deduced from the magnetic data themselves.

The energy differences between the ground and next highest states are 255 cm^{-1} and $\sim 400 \text{ cm}^{-1}$ for Eu^{+3} and Am^{+3} respectively; 932 and 1304 cm^{-1} for Sm^{+3} and Pu^{+3} , to give best agreement with the experimental data.

Consequently, in equation (1), $x = 365/T$ for Eu^{+3} , $570/T$ for americium. In (2), $y = 191/T$ for samarium, $267/T$ for plutonium.

The multiplet intervals are somewhat greater for the heavier elements, as is to be expected. Experimentally Z effective for americium and plutonium appears to be about 35-36.

The temperature dependence of susceptibility implied by equations (1) and (2) is strikingly different from the usual Curie or Curie-Weiss relationship, and the fact that Pu^{+3} and Sm^{+3} on the one hand and Am^{+3} and Eu^{+3} on the other deviate in the same way from the usual Curie-Weiss temperature dependence is a strong indication that the temperature deviations of analogous ions arise from the same basic cause. It is to be expected then that the J multiplet intervals of the lower states in Pu^{+3} and Am^{+3} are comparable to KT .

However, since the actual susceptibilities of solid compounds of Sm^{+3} , Eu^{+3} , Pu^{+3} and Am^{+3} deviate appreciably (especially in the heavy elements) from the relationships given by (1) and (2) which are based on the simple "field free" ion picture, the actual energy and scheme in the solid may deviate substantially from that given by the simple theory. In fact, the effect of the electrostatic fields of neighboring ions, and of the resonance interaction between neighboring paramagnetic ions would be expected to produce such deviations.

It has seemed desirable therefore to seek other evidence for the energy levels in the compounds of these elements and a program to that purpose has been initiated, of which the present work is a part. Besides magnetic measurements, heat capacity measurements,⁴ absorption spectrum measurements,⁵ and magnetic resonance measurements, furnish evidence concerning the energy levels in solids. Low temperature heat capacity measurements on radioactive materials are technically difficult because of self-heating of the materials, and thus far have been carried out successfully only for compounds of relatively low activity (i.e., Np^{237} , half-life 2×10^6 years.⁶)

However, absorption spectra are readily obtained from relatively small amounts of radioactive materials, the only technical difficulty being that highly hydrated compounds (such as salts of the type $M_2(SO_4)_3 \cdot 8H_2O$ or $MCl_3 \cdot 6H_2O$) which are desirable because they minimize the electrostatic and resonance perturbations are decomposed rather rapidly by the alpha particles. For that reason the present investigations were made on the stable anhydrous trifluorides of plutonium.

Absorption spectra, in contrast to emission spectra, offer the technical advantage that at sufficiently low temperatures all lines correspond to transitions from the lowest, or ground, state to some higher state. Furthermore, in principle at least, low lying levels may be detected by promoting the population of these levels (by raising the temperature) with the appearance of new lines corresponding to transitions from the new levels. In practice difficulties arise because of coupling with lattice vibrations, of smearing of the lines due to the rapidly varying crystal fields at higher temperatures, and perhaps because of variations in transition probabilities due to overall expansion of the crystal lattice.

Nevertheless, this method of detecting low lying levels in crystals was applied with seeming success by Spedding and his collaborators to $SmCl_3 \cdot 6H_2O$, etc.⁷

It was the intention of the present work to apply this technique to the trifluorides of plutonium, americium, samarium and europium in pure crystals and in solid solution in other compounds.

If it is indeed true that the ground states are the same for Pu^{+3} and Sm^{+3} on the one hand and of Eu^{+3} and Am^{+3} on the other, then this fact would be expected to be evident in systematic correlations between their absorption spectra. Because, in general, the transition probabilities in the transuranium elements are some ten to one hundred times as great as in the rare earths, and also because the Stark splitting by crystalline fields would be expected to be different in the two series of elements, the correlations probably would not be obvious.

Nevertheless, for the simple field free case the ratios of corresponding J multiplet intervals should be the same in Pu^{+3} and Sm^{+3} and in Eu^{+3} and Am^{+3} . Again, for corresponding transitions, the change in multiplicity induced by a given change in symmetry of the ionic environment should be the same for the analogous pairs of ions mentioned above.

Unfortunately, in the present work many technical difficulties prevented the achievement of more than a beginning on these problems. For example, it was found that SmF_3 , as prepared by the sublimation method described below, is of the orthorhombic crystalline type whereas PuF_3 is hexagonal. Not enough is known of these crystals to investigate their site symmetries. However, since the compounds form two different crystalline types, there seems little likelihood that the correlations hoped for could be observed.

II. EXPERIMENTAL

The rare earth and actinide fluorides precipitate from aqueous solution in a form unsuitable for absorption spectra measurements. The particles are so small that incident light is mainly reflected by the many surfaces rather than being transmitted. It was found that if the particles were immersed in a substance of high refractive index, scattering of light was greatly reduced, i.e., carbon disulfide renders powdered NdF_3 transparent. The first approach was to look for a substance which had a high refractive index, which would remain transparent at liquid nitrogen temperatures, and which would be transparent over a great portion of the spectral region.

Many compounds and solutions were tried. The best results were obtained from glycerol which had been saturated with lithium iodide. This solution has an index of refraction of 1.54 and forms a transparent glass at liquid nitrogen temperature. Excellent low temperature pictures were obtained from NdF_3 dispersed in this medium. Attempts to photograph PuF_3 by this method met with failure. The glycerol solution is apparently of little value for substances whose refractive index is as high as the 1.68 value reported for PuF_3 .⁸

The possibility of fusing PuF_3 into a transparent mass was next tried. In order to prevent formation of the oxide the heating was done in a vacuum system. The PuF_3 was contained in a platinum liner 2-1/2 cm long x 1 mm in diameter. It was hoped that the upper unheated section of the liner would condense any PuF_3 vapor. The liner was inserted into a beryllium oxide crucible 1-1/2 cm long with a 4 mm

outside diameter and a 1-1/2 mm inside diameter. The crucible with liner was inserted into a heating coil made of 35 mil tantalum wire, which was fused to 80 mil tungsten leads. These leads ran outside the vacuum system through a glass-to-metal seal. The current through the heating coil was controlled by means of a variac and a transformer.⁹

Throughout the run the temperature of the heating coil was increased very slowly. During this time the vacuum on the system was kept lower than 2×10^{-5} mm Hg as determined by an ionization gauge. The temperature was estimated by rough optical pyrometer readings. When it was felt the sample had been taken to a temperature of greater than 1600° C the current was shut off. Inspection of the liner revealed that the sample had completely sublimed onto the walls in the upper cooler region. The sublimate appeared very transparent and was transferred to a quartz plate for photographing. The sample was held to the plate by Duco cement which had been diluted with acetone. (The sample was verified to be PuF_3 by x-ray diffraction analysis.)

The optical arrangement used is described by Stover and Conway.¹⁰ The spectrograph used was a 21-foot, 15,000 line per inch Wadsworth mounted grating manufactured by Jarrell-Ash Company. Exposure times of from 1 to 6 minutes were required. The spectral region from 2500-9000 Å was investigated. No method of determining exact sample thickness could be devised. By focusing upon the top and bottom of the crystal with a calibrated depth focusing microscope a value of 90 ± 20 microns was obtained. In view of the uncertainty in sample thickness, calculation of absolute intensities seemed of little value.

In order to extend the observations in the ultraviolet direction, construction of a dewar with quartz windows was undertaken. Attempts to make a vacuum-tight seal of optical quartz to pyrex were unsuccessful. The next approach tried was the use of a gasket to hold a quartz window to a metallic dewar. After many gaskets had failed, a suggestion of Eugene V. Clark led to the trial of silicone rubbers produced by Dow Corning Corporation. It was found that a vacuum-tight seal was maintained at liquid nitrogen temperatures by a gasket of uncured Silastic No. 6-126 silicone rubber. The gasket is prepared simply by applying a coating of the rubber over the area where the window contacts the dewar and evacuating. Even though the rubber does not remain flexible at these temperatures the gasket is so thin that its contraction is not enough to break the seal. (The uncured rubber has the consistency of soft putty.) Using a stainless steel dewar with quartz windows, a water cooled hydrogen ultraviolet source and Eastman Kodak U.V. Sensitive Type 103-0 plates the spectral range was extended to 2500 Angstroms.

Even though many lines were observed at 77° K, no line absorption was found at room temperature. It was felt that this was not observed because of the small size of the sample. So much light was being transmitted around the sample and into the spectrograph that a high background resulted. This would render the broader, and hence less intense, room temperature absorption unobservable. It was found that room temperature absorption could be photographed by masking the lens so that the light passed through a hole 0.1 mm in diameter. It was

then possible to focus a pinpoint of light comparable to the sample size upon the sample. In this manner room temperature absorption was obtained.

Even at 77° K the lines obtained were so broad that it was difficult to locate the center of the line by visual observation. Absorption maxima were determined from densitometer recordings made on each line. Scratch marks were drawn from standard iron arc spectra so as to extend over the exposure of the PuF_3 spectrum. These markings also appeared on the densitometer recordings. The wave length of the absorption line was obtained by measuring the distance from the scratch mark to the line center. Each line was measured at least twice by this method. Most of the lines had a mean deviation of less than one Angstrom. The others could all be grouped into two classifications: those with mean deviation between one and two Angstroms and those with mean deviation between two and three Angstroms. The classification of lines into these categories is an indication of the sharpness of the line.

III. RESULTS AND DISCUSSION

In Table I are reported the wave lengths, intensities and wave numbers of the absorption lines of PuF_3 at 77° K. The intensities were classified from the densitometer recordings by arbitrarily assigning the most shallow peak the value of 0 and the peak of greatest height the value of 10. Wave lengths not otherwise marked have a mean deviation of less than one Angstrom.

Table I. Wave lengths, intensities and wave numbers of PuF_3 at 77°K .

$\lambda(\text{\AA})$	I	$\bar{\nu}(\text{cm}^{-1})$	$\lambda(\text{\AA})$	I	$\bar{\nu}(\text{cm}^{-1})$
^a 2686.5	1	^a 37212	4018.3	3	24879
^b 2750.6	2	^b 36345	4056.0	4	24648
2818.2	1	35473	^b 4120.6	5	^b 24261
3485.1	2	28685	^b 4140.9	6	^b 24142
3581.2	4	27916	4155.8	5	24056
3597.0	4	27793	^b 4196.6	3	^b 23822
3608.9	5	27701	4208.5	4	23755
3622.0	5	27601	^b 4214.6	3	^b 23720
3632.1	5	27524	^b 4223.4	3	^b 23671
3634.6	4	27505	4234.9	2	23607
3639.4	3	27469	4242.4	3	23565
3709.3	3	26952	4250.1	2	23522
^a 3867.3	3	^a 25850	4276.5	2	23377
3908.1	4	25681	4283.4	2	23339
3916.3	4	25527	^b 4291.2	2	^b 23297
3926.8	3	25459	4307.7	2	23208
3933.8	3	25413	4328.0	2	23099
3945.3	2	25339	4362.0	3	22919
3961.0	2	25239	4378.6	1	22832
3974.6	2	25153	4392.2	1	22761
3982.5	1	25103	4400.0	1	22721
3995.3	2	25022	4408.9	0	22675

Table I, cont.

$\lambda(\text{\AA})$	I	$\bar{\nu}(\text{cm}^{-1})$	$\lambda(\text{\AA})$	I	$\bar{\nu}(\text{cm}^{-1})$
4428.2	2	22576	5830.0	1	17148
4448.2	1	22475	5857.7	1	17067
4478.2	1	22324	5869.9	2	17031
4503.9	2	22198	5887.1	3	16982
4510.4	5	22165	5928.6	3	16863
4537.2	4	22034	5947.4	3	16809
4560.0	5	21924	^a 5994.9	2	^a 16676
4696.1	2	21288	6052.2	2	16518
4756.6	3	21018	^b 6085.8	1	^b 16427
4762.8	2	20990	6114.6	1	16350
4773.5	4	20943	6127.4	0	16316
4782.1	3	20905	6209.9	3	16099
4784.3	2	20896	6212.1	3	16093
4789.0	1	20875	6524.6	4	15322
4979.0	4	20079	6541.3	4	15283
4982.5	5	20065	6555.5	4	15250
4995.1	7	20014	6565.1	4	15228
4998.7	4	20000	6588.7	5	15173
^a 5238.6	5	^a 19084	7265.5	1	13860
^b 5297.3	9	^b 18872	7561.9	2	13320
^a 5367.4	10	^a 18626	7608.6	2	13139
^b 5427.5	8	^b 18420	^b 7657.7	3	^b 13055
A band of intensity 7 extends			7808.3	8	12803
from 5455 \AA to 5752 \AA .			7848.5	8	12737

Table I, cont.

$\lambda(\text{\AA})$	I	$\bar{\nu}(\text{cm}^{-1})$	$\lambda(\text{\AA})$	I	$\bar{\nu}(\text{cm}^{-1})$
7892.0	8	12668	8239.3	5	12133
^b 8143.2	5	^b 12277	8253.1	5	12113
8175.0	5	12229			

^aThese lines have a mean deviation between 2 and 3 Angstroms.

^bThese lines have a mean deviation between 1 and 2 Angstroms.

Room temperature spectra were also recorded in the hope of finding temperature dependent lines. The absorption was so broad, however, that it was impossible to resolve single lines. Lack of time prevented the investigation of intermediate temperatures. The broadness of the absorption spectrum of PuF_3 is in agreement with effects observed by Sancier and Freed.¹¹ In work on U^{+4} in three different environments they observe a greater dependence of the spectrum on environment than is observed for the lanthanide analog Pr^{+3} . This indicates the interaction of the fields of the environment with the ion is greater in the actinide case. Although they make no room temperature observations, one would expect this increased field interaction to cause room temperature absorption to be broader in the actinide case. Comparison of PuF_3 spectrum with SmF_3 spectrum, observed in this laboratory, substantiates this. Room temperature photographs of SmF_3 exhibit reasonably sharp absorption lines.

Wave number differences between all of the lines were determined. No difference of greater than $4,000 \text{ cm}^{-1}$ was obtained. A plot of line differences against a unit scale allowed constant differences to be recognized by a sudden change in slope. The following constant differences were observed:

85.3 (9)	507.1 (8)	1271.5 (7)	2320.1 (6)
267.9 (7)	719.1 (7)	1539.5 (6)	2579.4 (8)
376.2 (8)	1044.6 (7)	1581.5 (8)	2765.2 (9)
389.9 (9)			3747.1 (7)

The numbers in parentheses indicate the number of differences which fall within 2.5 wave numbers of the constant differences. The constant differences are mean values.

The statistical significance of these constant differences is somewhat doubtful, since the precision of measurement of many lines is no better than $\pm 1 \text{ \AA}$. However, if the energy levels in the crystal are situated in such a manner that equal distances occur between levels, at least some of these constant differences will represent those distances.

A relationship between the differences 267.9, 1044.6, 1271.5 and 1539.5 should be indicated. The sum of these differences is found to equal other constant differences:

$1044.6 + 1271.5 = 2316.1$	which is to be compared with	2320.1
$1539.5 + 1044.6 = 2584.1$	"	2579.4
$267.9 + 1271.5 = 1539.4$	"	1539.5
$267.9 + 1271.5 + 1044.6 = 2584.0$	"	2579.4.

If these differences are all statistically significant, then four levels which are separated by 267.9, 1271.5 and 1044.6 cm^{-1} must recur. The magnitude of the splitting eliminates the possibility of this being a split ground state since Boltzmann's distribution does not allow appreciable occupancy of levels 1000 cm^{-1} above the ground state at 77° K.

The question of low lying levels remains unanswered. If they exist they will be separated by one or more of the differences 85.3, 267.9, 376.2, 389.9, 507.1 or 719.1

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