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Publication Date
1965-07-01

UCRL-16292

University of California

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Berkeley, California

Submitted to Journal of Chemical Physics UCRL-16292

UNIVERSITY OF CALIFORNIA

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AEC Contract No. W-7405-eng-48

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ABSTRACT

Polarized absorbtion spectra and some Zeeman effect measurements in the region of 20,000-32,000 cm^{-1} are reported for Pu^{3+} in $LaCl_3$. A total of 25 J-levels was fitted with an rms deviation of 100 cm^{-1} with the parameters values $E^0 = 14930.5$, $E^1 = 3726.7$, $E^2 = 14.99$, $E^3 = 350.18$, $\zeta = 2260.2$, $\alpha = 36.5$, $\beta = -1171$. The large values of α and β indicate that configuration interaction effects cannot be neglected in an analysis of the spectra of the higher actinides.

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Lämmermann and Conway[1] have reported work on Pu^{3+} in $LaCl_3$ for the multiplets from 0-20,000 cm^{-1} and have treated these using 5f hydrogenic wave functions. Above 20,000 cm^{-1} the density of levels is high and there was no correlation with the simple hydrogenic calculation. Since it was not possible to vary all 4 parameters simultaneously, no attempt was made to analyze the data beyond 20,000 cm^{-1}.

Recent studies of configuration interaction effects in rare earth ions[2-4] have prompted an interest in this problem to determine values of the configuration interaction parameters for the actinides. At present Pu^{3+} is the only trivalent actinide on which there is sufficient data for such a calculation.

The calculation procedures described previously[3,4] were used to fit 12 levels below 20,000 cm^{-1} with the five parameters E^0, E^1, E^2, E^3,

and ζ (Calc. I). The rms deviation[5] was 325 cm^{-1} compared with a value of about 500 cm^{-1} for the simple hydrogenic calculation. The addition of the configuration interaction parameters α and β [6] (Calc. II) further reduced this deviation to 103 cm^{-1}. The good agreement and large shifts in many of the calculated levels prompted us to extend this analysis.

The additional data for Pu^{3+} in $LaCl_3$ given in Table I are derived from plates taken previously by Lämmermann. New assignments were made from consideration of Zeeman splittings and correlations between experimental and calculated energies. Individual J-level assignments are discussed below. The only new assignment below 20,000 cm^{-1} is Group K. The crystal field quantum numbers μ in Table I are related to the $\bar{\mu}$ used previously[1] by: $\mu = \bar{\mu}$ for $\mu = 3/2$; $\bar{\mu} = 5/2$ is $\mu = 1/2$; $\bar{\mu} = 1/2$ is $\mu = 5/2$. The ground state is $^6H_{5/2}$ $\mu = 1/2$. This level is labeled I. The first excited level at 13 cm^{-1} is $\mu = 3/2$ and is designated II.

Groups I and K: Lämmermann and Conway reported two groups of levels in the region of 16-17,000 cm^{-1}, I at 15,963 and K at 16,395. Group I consists of only 2 lines and is obviously $J = 3/2$. Group K consists of at least 6 lines, 3 with $\mu = 3/2$. This suggests $15/2 \leq J \leq 19/2$. This group was not previously assigned because the calculation put $J = 15/2$ below $J = 3/2$ and no value of $\chi = \zeta/F_2$ could invert this order. However, the free variation of 5 parameters reversed the previous order and the agreement between theory and experiment is now satisfactory. The $J = 9/2$ which was previously considered as a possible assignment for K is now calculated to be 800 cm^{-1} above Group K.

Group 20,725: This is a well isolated group containing 3 $\mu = 5/2$ levels which implies $J \geq 17/2$. No other levels are calculated in this

region and the assignment of J = 17/2 is unambiguous.

Group 21,621: Calc. II put two levels in this region, J = 5/2 at 21,855 and J = 9/2 at 21,800. The perpendicular splitting of the σπ line at 21,644.6 indicates J ≥ 7/2. Furthermore the $s_\perp$ = 5.38 of the σ line at 21,601.1 can be explained only by a transition from II to the μ = 1/2 level of J = 9/2.

Group 24,697: Calc. II indicates a J = 11/2 level in this region with a g value of 0.9. This would give a maximum $s_\perp$ = 4.8 which is consistent with the $s_\perp$ = 4.15 observed for the lowest σπ line of this group.

Group 25,598: Levels with J = 3/2, 7/2, and 17/2 and g ~1 are predicted in this region by Calc. II. The splitting factor $s_\perp$ = 13.5 for the lower level of this group is much larger than would be expected for any level with g of 1. Some mechanism involving the interaction in the magnetic field of two nearly degenerate crystal field components must be invoked to get such a large splitting factor. Regardless of its origin, it is clear that a very high J-value must be involved. Consequently this group has been assigned J = 17/2.

Group 28,171: Two J values are calculated between 28,000 and 28,300 cm^{-1}. The J = 1/2 level should not be seen except by a transition from II, the excited state 13 cm^{-1} above ground. One of the weak σ lines in this group could be a transition to the J = 1/2, but the rest of the levels must belong to J = 11/2.

Group 28,558: The next level, J = 17/2, is separated by 200 cm^{-1} from a group containing a J = 5/2 and 2 J's of 9/2. The large perpendicular splitting of one line rules out the J = 9/2 and 5/2 as possible assignments.

Group 29,461: The possible assignments for this group are J = 3/2 and 15/2. The two $\sigma\pi$ lines indicate a J greater than 3/2 is present. The perpendicular splitting is reasonable for the J = 15/2 but it is not distinctive. Certainly, the J = 15/2 is present in this group, but one or more of the lines could belong to the J = 3/2.

Groups 22,054, 22,347, 22,614, 22,416, 25,170, 26923: These groups were assigned to J values of 5/2, 15/2, 7/2, 5/2, 13/2, 5/2, respectively because of the good agreement with calculations.

RESULTS

Using 7 parameters and 25 levels, the fit remained essentially unchanged at a deviation of 100 cm^{-1} (Calc. III). The parameter values given in Table II are slightly different from those obtained by fitting only the levels below 20,000 cm^{-1} (Calc. II). The results of Calculations I and III are compared with the experimental data in Fig. 1. The broad hatched areas indicate groups of lines which are unassigned. In most cases there are several J-levels calculated in these regions. The experimental groups show no obvious break down into subgroups. This is probably due to the effect of J-mixing. It is clear from Fig. 1 that for the higher levels the parameters α and β have considerable effect on both the position and order of the levels. Additional configuration interaction parameters could probably improve the fit obtained here, but the value of such an improvement would be questionable because of the uncertainty in the centers of gravity used and the unknown effects of J-mixing. These uncertainties probably account for at least 50 cm^{-1} of the present deviation. The results

of Calc. III are tabulated in Table III. Only those components with coefficients greater than 0.3 are included in the eigenvectors. The notation is that of Neilson and Koster.[7]

The fit of 100 cm^{-1} for Pu^{3+} can be compared to one of 80-90 cm^{-1} resulting from similar calculations for Nd^{3+} and Er^{3+}.[4] The α and β values of 36.5 and -1171 for Pu^{3+} are much larger than the $\alpha = 1.24$ and $\beta = -148$ for Nd^{3+} and the $\alpha = 17.9$ and $\beta = -743$ for Er^{3+}. This is an indication that configuration interaction effects are much more important in the actinides than in the rare earths. If α and β increase as rapidly across the actinide series as they seem to across the rare earth series, they must certainly be included in any interpretation of the spectra of the higher actinides.

ACKNOWLEDGMENTS

We wish to thank Dr. Brian Judd for his helpful discussion.

This work was performed under the auspices of the United States Atomic Energy Commission.

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2. K. Rajnak and B. G. Wybourne, Phys. Rev. 132, 280 (1963).

3. K. Rajnak, J. Opt. Soc. Am. 55, 126 (1965).

4. K. Rajnak, J. Chem. Phys. 43, 847 (1965).

5. The rms deviation was defined as $(\Sigma_i \Delta_i^2/(N-K))^{1/2}$ where Δ is the deviation of the i^{th} level, N is the number of levels, and K, the number of variable parameters.

6. α is the coefficient of the term L(L+1) in the Hamiltonian and β is the coefficient of $G(G_2)$.

7. C. W. Nielson and George F. Koster, Spectroscopic Coefficients for the p^n, d^n, and f^n Configurations. (The M.I.T. Press, Massachusetts Institute of Technology, Cambridge, Massachusetts, 1963).

Table I. Absorbtion lines of Pu^{3+} in $LaCl_3$.

Center[a]	Energy (cm^{-1})	P[b]	I[c]	Lower level	$s_{\|}$	$s_{\perp}$	μ	Remarks
	20624.7	σπ	5				5/2	
	20671	π	T[d]	II				
	20684.4	σ	6	I			3/2	Broad
	20730	π	T	II				
20755	20742.4	σ	4	I			3/2	
	20792.9	σπ	4				5/2	Not temperature dependent
	20805.4	π	T?					
	20810.6	π	4					
	20825.3	σπ	4				5/2	
	21601.1	σ	3	II		5.38	1/2	
	21631.5	σ	T	II				⊥ splitting not measureable
21651	21644.6	σπ	10	I		2.92		
	21655.8		0					
	21695.3	π	T	II				
	21708.5	σ	8	I			3/2	
	21751.0		ν[e]					
	21776.6		ν					
	21799.9		ν					
	21838.4		ν					
	21867.1		ν					
	21883.8		ν					
	21896.3		ν					
	22936.3	σ	4					
22054	22070.8	π	10					
	22303.7	σ	2					⊥ splitting not measureable
	22309.7	π	2					⊥ splitting not measureable
22347	22365	π	T	II				
	22378.1	σ	2	I			3/2	
	22395.1	π	6			4.3		

Table I. Continued.

Center[a]	Energy (cm^{-1})	P[b]	I[c]	Lower level	$s_{\|}$	$s_{\perp}$	μ	Remarks
	22499		ν					
	22525.3		ν					
	22534.5		ν					
	22540.6		ν					
	22600.4	π	7					broad
22614	22627.8	σ	7					
	22956.7	σ	10					
	23083.4	σπ	10					
	23093.0		1					
	23172.2	π	T					
	23184.6	σ	8					
	23232	σ	T					
	23245.5	σπ	10				5/2	
	23280.1	σ	1			4.15		
	23293.7	σπ	7			2.92		
	23342.1	σ?	1					
	23372.7	σπ?	9					
	23382.5		7					
	23407.1		1					
	23612.2	σ	4					
	23650	σ						
	23653	π						
	23663.1	π	T	II	2	5.23		
	23672.6	π	1					
	23686.6	σ	1					
	23705.7	σ	5					
	23711.9	π	5					
	23736.1	σπ	1					
	23772.8	σπ	3					
	23848	π	2					
	23864.8	π	3					

Table I. Continued.

Center[a]	Energy (cm^{-1})	P[b]	I[c]	Lower level	$s_{\|}$	$s_{\perp}$	μ	Remarks
	24087.6	$\sigma\pi$	10					
24146	24159.4	σ	2					
	24191.0	$\sigma\pi$	6					
	24590.9	$\sigma\pi$	3					
	24680	π	T	II	2	4.15		
24697	24693.5	σ	0	I	2.0		3/2	
	24807.5	$\sigma\pi$	0					Broadened in $H_{\perp}$
	25051.8	π	T	II	4.2			
	25064.4	σ	6	I	3.8		3/2	
	25121	σ	T					
25170	25133.7	π	4		0	4.85		
	25187.5	$\pi, 1/2\sigma$	10			4.85		Could be 2 lines
	25219.9	σ	3					
	25246.0	σ	5					
	25512.1	σ	2			13.5		
	25530	π	T	II				
	25542.7	σ	4	I			3/2	
25598	25545	π						$\|$ splitting not measurable
	25606.2	π	3					
	25627.2	π?	3					
	25700.3	$\sigma\pi$	4					broad
	26003.8	π	5					broad
	26043.8	π	5					broad
	26125.4	$\sigma\pi$	5					broad
	26315	σ	T	II				broad
	26328.4	$\sigma\pi$	7	I			5/2	broad
	26390.3	$\pi, \frac{1}{4}\sigma$	10		6.2			
	26408.4	π	T	II	3.8			
	26421.6	σ	5	I	3.8		3/2	

Table I. Continued.

Center[a]	Energy (cm^{-1})	P[b]	I[c]	Lower level	$s_{\parallel}$	$s_{\perp}$	μ	Remarks
26923	26908.8	$\pi,\frac{1}{4}\sigma$						sharp
	26937.8	$\sigma,\frac{1}{4}\pi$			2.5			
	27070.6	π	0					
	27086.7	π	5					
	27091.8	π	4					could be 1 or 2 lines
	27097.0	π	3					
	27124.9	$\sigma\pi$	9					
	27135.2	σ	6		3.8			
	27151	π	T		3.8			
	27161.0	σ	3					
	27231.3	$\pi,\frac{1}{2}\sigma$	10					possibly 2 lines, one σ and one π
	27244.6	π	T	II			3/2	
	27507.0	σ	10		2.5			
	27974.2	π	T					
	28125.3	σ	8					
	28129	σ	T	II				
28171	28132.4	$\sigma\pi$	10	I		2.46	5/2	
	28188	π		II				
	28200.7	σ	7	I	6.66		3/2	
	28219.8	$\sigma\pi$	9					
	28263.7	π?	1					
	28283.7	σ?	1					
	28506.6	σ	5					
	28515.2	$\sigma\pi$	7			7.54	5/2	
28558	28543	σ	T	II	5			
	28555.9	π	8		4.3			
	28571.4	σ	4					
	28592.7	σ	9					not temperature dependent
	28605.8	$\pi,\frac{1}{2}\sigma$	10		13			

Table I. Continued.

Center[a]	Energy (cm^{-1})	P[b]	I[c]	Lower level	$s_{\parallel}$	$s_{\perp}$	μ	Remarks
	28679.6	π	0					
	28701.0	$\sigma\pi$	1					
	28758.0	σ	1					
	28851.8	π?	0					
	29085	π	T	II				
	29097.8	σ	9	I	3.3		3/2	
	29104	σ	T	II				
	29210.9	π	T	II				
	29347.2	$\sigma\pi$	6		3.3	2.0	5/2	
29461	29445.8		0					
	29463.1	σ	4					
	29496.2		0					
	29552.0	$\sigma\pi$	6					broad
	29813	$\sigma,\frac{1}{2}\pi$	T		4.6			
	29826.1	$\sigma,\frac{1}{4}\pi$	9		4.6			
	29885.0	$\sigma,\frac{1}{4}\pi$	6		5.4			
	29945.0	$\pi,\frac{1}{2}\sigma$	9					
	29995	π	T					
	30007.9	σ	2					
	30349	σ	T	II				
	30362.4	$\sigma\pi$	7	I	4.6		5/2	
	30459	σ	T	II	3.6			
	30472.5	$\sigma\pi$	3	I	4.6	5.0	5/2	
	30503.2	$\sigma\pi$	3					
	30519.0	$\sigma\pi$	3					
	30523	σ	T		3.6			
	30551	π	T	II	3.6			
	30563.8	σ	7	I	6.0		3/2	
	30583	π	T?					

Table I. Continued.

Center[a]	Energy (cm^{-1})	P[b]	I[c]	Lower level	$s_{\parallel}$	$s_{\perp}$	μ	Remarks
	30620	π	T?					character different from other groups. Could be vibronic
	30638.8	$\sigma\pi$	4					
	30823	$\sigma\pi$						
	30835	σ						
	31205.9	σ	T	II				
	31213	σ						
	31219.5	$\sigma\pi$	4				5/2	
	31225.4	$\sigma\pi$	3					$\parallel$ splitting not measurable
	31231.2	$\sigma\pi$	2					
	31578.4	σ	T	II				
	31591.4	$\pi,\frac{1}{2}\sigma$	7					
	31648	σ	T					
	31669.5	π	T					
	31681.5	σ	3					
	31712.7	σ	7					
	31783.2	σ	4					broad
	31816.6	$\sigma\pi$	0					broad
	31838.9	$\sigma\pi$	0					broad

[a] Center of gravity relative to lowest level of $^6H_{5/2}$; computed as the average of those lines not obviously arising from II. Center of $^6H_{5/2}$ = 29.7 cm^{-1}.

[b] Polarization. Entries such as $\sigma,\frac{1}{2}\pi$ indicate that the π component was only half as intense as the σ component.

[c] Intensity in 1% crystal.

[d] Temperature dependent.

[e] Probably vibronic. Broad, unpolarized, not temperature dependent, but highly concentration dependent.

Table II. Parameter values for Pu^{3+} in $LaCl_3$ (cm^{-1}).

	Hydrogenic[a]	I	II	III
E^0		14601.4	14890.9	14930.5
E^1	3677.29	3810.99	3753.77	3726.70
E^2	18.68	21.62	14.93	14.99
E^3	358.32	316.67	354.08	350.18
ζ	2290	2381.34	2254.3	2260.2
α			38.61	36.50
β			-1211	-1171
No. levels		12	12	25
σ (cm^{-1})	500	325	103	100

[a]From Reference 1.

Table III. Energy levels, eigenvectors and g values for Pu^{3+} in $LaCl_3$.

J	E_{calc}	E_{exp}	ΔE	Eigenvector[a]	g
5/2	-82	0	-82	$0.81\,^6H$ $-0.31\,^4G1$ $-0.38\,^4G4$	0.420
7/2	3237	3190	47	$0.90\,^6H$ $-0.31\,^4G4$	0.865
9/2	6191	6038	153	$0.93\,^6H$	1.070
5/2	6802			$-0.69\,^6F$ $+0.32\,^6H$ $+0.35\,^4F3$	1.141
3/2	6910			$0.84\,^6F$ $-0.33\,^4F3$	0.977
1/2	7215			$0.91\,^6F$	-0.536
11/2	8703	8663	40	$0.91\,^6H$	1.177
7/2	9604	9557	47	$-0.82\,^6F$ $+0.34\,^4F3$	1.322
13/2	10822	10938	-116	$0.86\,^6H$ $+0.37\,^4I3$	1.232
15/2	12270			$0.72\,^6H$ $+0.48\,^4I3$	1.227
9/2	12275			$-0.87\,^6F$	1.381
5/2	13670	13596	74	$-0.60\,^6F$ $-0.36\,^6H$ $-0.31\,^4G4$	0.966
11/2	14783	14828	-45	$-0.86\,^6F$ $+0.31\,^4G2$	1.400
3/2	15924	15963	-39	$0.43\,^6F$ $-0.32\,^4F1$ $+0.73\,^4F3$ $+0.33\,^4F4$	0.568
15/2	16314	16395	-81	$-0.43\,^6H$ $-0.30\,^4L$ $+0.67\,^4M$ $-0.38\,^2L3$	0.974
5/2	17075			$0.54\,^6P$ $+0.52\,^4P2$	1.403
9/2	17229			$0.33\,^4I1$ $+0.74\,^4I3$ $+0.31\,^2H6$	0.820
13/2	17734			$-0.35\,^6H$ $-0.42\,^4K1$ $-0.66\,^4L$	0.940
11/2	17760			$-0.45\,^4I3$ $+0.63\,^4K1$	0.908
7/2	18223	18257	-34	$-0.40\,^6F$ $-0.30\,^4G1$ $+0.35\,^4G2$ $-0.48\,^4G4$	1.053
3/2	19290	19350	-60	$-0.76\,^6P$ $-0.55\,^4P2$	2.031

Table III. Continued.

J	E_{calc}	E_{exp}	ΔE	Eigenvector[a]	g
1/2	20071			$-0.33\,^6F\ -0.31\,^4D3\ +0.31\,^2P1\ -0.46\,^2P3\ +0.58\,^2P4$	0.435
17/2	20614	20725	-111	$0.45\,^4L\ -0.79\,^4M$	1.012
9/2	21740	21621	119	$0.47\,^4G2\ -0.48\,^4G4\ -0.43\,^4H1\ -0.30\,^4H3$	1.108
5/2	21854	22024	-170	$0.60\,^4F3\ +0.36\,^4G3$	0.915
15/2	22416	22317	99	$0.48\,^4K1\ +0.58\,^4L\ +0.45\,^4M$	1.007
7/2	22634	22584	50	$0.37\,^4G3\ -0.41\,^4H1\ -0.44\,^4H3\ -0.36\,^2G5$	0.875
9/2	23163			$-0.33\,^6F\ -0.34\,^4F2\ -0.30\,^4G2$	1.069
7/2	23433			$0.74\,^6P\ +0.34\,^4D1\ +0.32\,^4D2$	1.526
11/2	23483			$0.400\,^4G2\ -0.56\,^4H1$	1.123
19/2	23570			$0.42\,^4L\ -0.80\,^4M\ -0.36\,^2N$	1.068
13/2	23724			$-0.39\,^4I3\ +0.55\,^4K1\ -0.58\,^4L$	0.953
15/2	23920			$0.37\,^6H\ -0.35\,^4I3\ -0.46\,^4L\ +0.34\,^4M\ +0.38\,^2K5$	1.068
5/2	24068	24116	-48	$-0.58\,^6P\ +0.40\,^4D2$	1.417
11/2	24631	24668	-37	$0.68\,^4I3\ +0.51\,^4K1$	0.922
13/2	25198	25141	57	$0.52\,^4I3\ +0.37\,^4K1\ -0.43\,^2K5$	1.024
21/2	25202			$0.77\,^4M\ +0.57\,^2N$	1.098
3/2	25473			$0.33\,^4P1\ -0.53\,^4D2\ +0.41\,^4D3$	1.316
17/2	25694	25568	126	$0.67\,^4L\ +0.32\,^4M\ -0.34\,^2L3\ +0.44\,^2M1$	1.038
7/2	25768			$-0.34\,^4F3\ +0.33\,^4G1\ +0.34\,^4G2\ -0.44\,^4G3$	1.069
13/2	26070			$-0.55\,^4H1\ -0.37\,^4H3\ +0.42\,^4I2$	1.129
9/2	26247			$0.30\,^4F3\ +0.33\,^4G4\ -0.46\,^4H3$	1.060
7/2	26310			$0.31\,^6F\ +0.47\,^4F3\ +0.45\,^4G2\ -0.36\,^4H1$	1.045

Table III. Continued.

J	E_{calc}	E_{exp}	ΔE	Eigenvector[a]	g
5/2	26906	26894	12	$-0.76\,^4G2 + 0.32\,^4G3$	0.708
11/2	27001			$-0.30\,^6F - 0.33\,^4H3 + 0.35\,^2I4$	1.114
7/2	27227			$-0.49\,^6P + 0.50\,^4D2 - 0.40\,^4D3 + 0.30\,^4F2$	1.405
3/2	27481			$-0.44\,^6P + 0.38\,^4P2 + 0.30\,^2P3 - 0.41\,^2P4$	1.489
1/2	28143			$0.81\,^4P2 - 0.33\,^4D3$	1.995
11/2	28217	28141	76	$0.31\,^4G2 - 0.53\,^4G4 + 0.34\,^4H3 + 0.39\,^2H6$	1.167
17/2	28415	28528	-113	$-0.72\,^4K1 - 0.43\,^4M - 0.37\,^2L3$	1.099
9/2	28614			$0.68\,^4F2 + 0.41\,^4H1$	1.170
5/2	28660			$-0.30\,^4D2 - 0.58\,^4G3 - 0.34\,^2F7$	0.939
9/2	28702			$-0.43\,^4G3 + 0.53\,^4I2 - 0.30\,^2H4 + 0.30\,^4H3$	1.001
15/2	29457	29431	26	$-0.70\,^4K1 + 0.52\,^4L$	1.054
3/2	29495			$0.34\,^4P1 - 0.37\,^4D3 - 0.46\,^4F2 - 0.31\,^2P3 + 0.49\,^2D3$	1.036
5/2	29844			$0.56\,^4F2 - 0.31\,^2F4$	1.113
15/2	30125			$0.65\,^4I2 + 0.45\,^2K2 + 0.35\,^2K3 + 0.34\,^2L2$	1.105
19/2	30430			$0.69\,^4L + 0.54\,^4M - 0.40\,^2M1$	1.106
9/2	30466			$-0.57\,^4F3 - 0.34\,^2G5 - 0.26\,^2N$	1.183
11/2	30833			$-0.70\,^4I2 + 0.42\,^2H4$	1.024
7/2	31652			$-0.36\,^4D3 - 0.67\,^4F2 - 0.30\,^2F5$	1.160
9/2	31790			$0.47\,^4H1 - 0.45\,^2H6$	1.006
19/2	32068			$0.44\,^4L + 0.84\,^2N$	1.004

[a]Only those components with coefficients greater than .3 are given. Notation is that of Nielson and Koster.[7]

FIGURE CAPTION

Fig. 1. Energy levels of Pu^{3+}. a) Fit with 5 parameters (Calc. I) b) Fit with 7 parameters (Calc. III). c) Experimental levels. Levels are labeled by 2J. Broad hatched areas indicate groups of experimental levels which were not included in the fit.

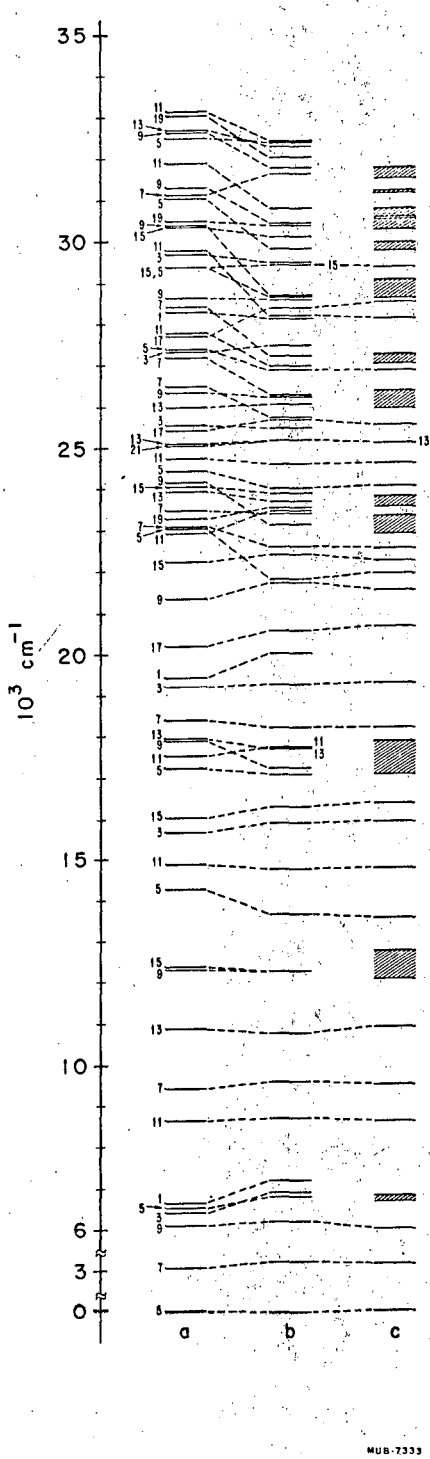

Fig. 1