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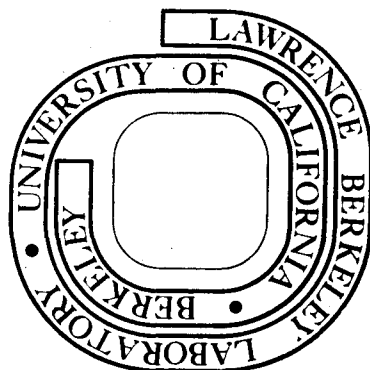
D. Brown, P. Lidster, B. Whittaker, and N. Edelstein

August 1975

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Contribution from the Nuclear Chemistry Division, Lawrence Berkeley Lab., University of California, Berkeley, California 94720 and from the Chemistry Division, Atomic Energy Research Establishment, Harwell, Oxon, England.

The Preparation and Spectral Properties
of Actinide (IV) Hexaiodocomplexes.

D. Brown, P. Lidster, B. Whittaker and N. Edelstein.

Hexaiodocomplexes of the type $R_2^1 M^{IV} I_6$ ($R^1 = NEt_4$ and NMe_3Ph ; $M^{IV} = Th, Pa$ and U) have been prepared by reactions in oxygen-free, anhydrous methyl cyanide. I.r. and Raman data are reported together with electronic spectral results for the protactinium (IV) and uranium(IV) complexes. Ligand field and spin-orbit coupling constants derived for the hexaiodoprotactinates (IV) are discussed in relationship to values obtained earlier for other hexahalogenoprotactinates (IV), $(NEt_4)_2 PaX_6$ ($X = F, Cl$ and Br).

As part of a detailed investigation of the trends in bonding of $5f^1$ compounds we have previously reported^{1,2} optical spectral data for octahedral compounds of the type $(NEt_4)_2 PaX_6$ ($X = F, Cl$ and Br). The trends in the ligand field parameters θ and Δ were explained qualitatively in terms of molecular orbital theory with large variations in σ bonding dominating the total ligand field splitting as the halide was varied, a feature which was also observed for certain hexahalogenouranates (V). Hexaiodo-complexes of tetravalent actinide elements have not been extensively studied^{3,4} and the single protactinium (IV) complex reported⁴, $(Ph_3MeAs)_2 PaI_6$, was only prepared on a very small scale and its physical properties were not examined.

We report now the preparation of new hexaiodocomplexes, $(\text{NEt}_4)_2\text{M}^{\text{IV}}\text{I}_6$ ($\text{M}^{\text{IV}} = \text{Th}, \text{Pa}$ and U) and $(\text{NMe}_3\text{Ph})_2\text{M}^{\text{IV}}\text{I}_6$ ($\text{M}^{\text{IV}} = \text{Pa}$ and U) together with details of their i.r., Raman and (Pa and U only) solid state electronic spectra, and X-ray powder diffraction results. The ligand field and spin-orbit coupling constants derived from the electronic spectra of the hexaiodoprotactinates (IV) are compared with those available for other octahedral $5f^1$ compounds.

EXPERIMENTAL

All reactions and sample preparation were performed in inert-atmosphere dry boxes (oxygen $< 20\text{ppm}$; $\text{H}_2\text{O} < 20\text{ppm}$) on account of the ready hydrolysis and oxidation (Pa) of the complexes and to afford protection against the radioactivity associated with ^{231}Pa .

Reagents Thorium³, protactinium⁴ and uranium³ tetraiodide were prepared according to the methods in the references cited. Commercially available NEt_4I and NMe_3PhI (B.D.H. Ltd.) were vacuum dried at room temperature (10^{-4}Torr) for several hours after which time no bands due to water were present in their i.r. spectra. Methyl cyanide was purified as described previously⁵, flushed with argon (99.999% purity) and stored in the inert-atmosphere glove boxes over freshly dried molecular sieves. It was passed down a column of new sieves immediately prior to use.

Hexaiodo-Complexes, $\text{R}_2\text{M}^{\text{IV}}\text{I}_6$ ($\text{R} = \text{NEt}_4$ or NMe_3Ph ; $\text{M}^{\text{IV}} = \text{Th}, \text{Pa}$ and U).

The appropriate tetraiodide (0.05 - 0.5g) was heated with the stoichiometric quantity (1:2 mole ratio) of tetraalkyl/aryl-ammonium iodide in anhydrous, oxygen-free methyl cyanide (0.2 - 1.0ml). The reaction mixture was cooled in ice, the crystalline product isolated by centrifugation, washed with ice-cold methyl cyanide and vacuum dried at room temperature. Yields were in the range 70-90%. Anal. Calc. for $(\text{NEt}_4)_2\text{ThI}_6$: Th, 18.51; I, 60.74%. Found: Th, 18.60; I 59.85%. Calc. for $(\text{NEt}_4)_2\text{UI}_6$: U, 18.89; I, 60.43%. Found: U 18.84; I 60.24%. Calc. for $(\text{NMe}_3\text{Ph})_2\text{UI}_6$; U, 18.72; I 59.86 Found: U, 18.75; I, 59.92%. The hexaiodoprotactinates (IV) which are isostructural

with their thorium and uranium analogues, were identified by X-ray powder diffraction analysis.

Analysis Thorium, uranium and iodide were determined as described previously³.

Physical Measurement Solid state transmission spectra (0.2 to 2.2 μ) were recorded using the technique described earlier⁶. I.r. and Raman spectra were recorded on a Perkin-Elmer 180 instrument and a Cary³ model 83 (488 nm argon-ion laser excitation) instrument, respectively, as described elsewhere⁷. X-ray powder diffraction samples were mounted in 0.3mm diameter Lindemann glass capillaries, which were sealed and coated with Bostikot⁸ to prevent accidental breakage. Films were recorded using a 19 cm Debye-Sherrer camera with nickel-filtered CuK_{α} radiation ($\lambda_{\text{m}}, 1.5418\text{\AA}$).

RESULTS AND DISCUSSION

The hexaiodocomplexes crystallise when the appropriate tetraiodide and alkyl/aryl-ammonium iodide are mixed in anhydrous, oxygen-free methyl cyanide. The products are extremely sensitive towards moisture, and for small-scale preparations (e.g. 50 mg of PaI_4) it is essential to pass previously purified methyl cyanide down a column of fresh molecular sieves in order to ensure complete absence of water. They are moderately soluble in methyl cyanide and insoluble in isopentane. The addition of aqueous ammonia results in the formation of hydrous oxides, the characteristic black protactinium (IV) hydroxide turning white within a few minutes in the presence of oxygen.

Partial X-ray powder diffraction data for $(\text{NEt}_4)_2\text{PaI}_6$ are listed in Table 1 together with visually estimated intensities: the thorium and uranium analogues are isostructural. The complexes appear to possess low symmetry and unit cells dimensions have not been obtained.

I.R. and Raman Spectra. Two infrared-active fundamentals, ν_3 (t_{1u}) and ν_4 (t_{1u}) and three Raman-active fundamentals, ν_1 (a_{1g}), ν_2 (e_g) and ν_5 (t_{2g}) are predicted by group theory for regular octahedral molecules of the type MX_6 belonging to

the point group O_h . The t_{2u} mode, ν_6 , is totally forbidden. Comparison of our spectra for the hexaiodo-complexes with those for hexafluoro-, hexachloro- and hexabromo-complexes of a variety of tetravalent actinide elements⁷ results in the assignments shown in Table 2. As indicated, Raman data could not be obtained for either $(NEt_4)_2PaI_6$, which exhibited strong fluorescence, or $(NEt_4)_2UI_6$, which decomposed in the laser beam. A definite assignment of ν_4 was not possible: a relatively weak, split absorption was observed around 60 cm^{-1} but this coincided with the probable position of the lattice mode and with a band observed in the spectra of thick mulls of NEt_4I . However, there is probably a contribution to this absorption from ν_4 since, on a mass basis (e.g. $\nu_4(I) = \sqrt{\frac{M(Cl)}{M(I)}} \cdot \nu_4(Cl)$ and $\nu_4(I) = \sqrt{\frac{M(Br)}{M(I)}} \cdot \nu_4(Br)$ with $\nu_4(Cl) = \text{ca } 110\text{ cm}^{-1}$ and $\nu_4(Br) = \text{ca } 81\text{ cm}^{-1}$) this mode would be expected between 55 and 65 cm^{-1} . The ν_2 mode was not observed in the Raman spectrum of $(NEt_4)_2ThI_6$. Although this mode was recently reported⁹ as a medium intensity band at 230 cm^{-1} for solid hexachlorouranates (IV) we have been unable to reproduce this and believe⁷ that the observation was incorrect. Our results confirmed those of Berringer et al¹⁰ who observed only ν_1 and ν_5 in the spectra of hexachloroneptunates (IV).

A band observed at 100 cm^{-1} in the Raman spectra of certain samples of $(NEt_4)_2ThI_6$ is believed to be due to hydrolysis products since it increases relative to other bands on exposure of the compound to moisture and is the only band present in the spectrum of an aqueous solution of $(NEt_4)_2ThI_6$. This band is therefore valuable in assessing the purity of a sample of this material. It is not possible at present to decide which of the two remaining bands (Table 2) should be assigned to ν_5 , and which to a lattice mode.

Electronic Spectra The solid-state spectra obtained for $(NEt_4)_2PaI_6$ are shown in Figures 1 and 2 and that for $(NEt_4)_2UI_6$ at 85K in Figure 3. The last has not been interpreted and will not be discussed in detail.

The measured band positions for $(NEt_4)_2PaI_6$ and $(NPhMe_3)_2PaI_6$ are provided in Table 3 together with assignments. The estimated accuracy of

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measurement is $\pm 10\text{\AA}$. Bands present in the spectra (Figures 1 and 2) at ca 1.68μ are associated with transitions within the tetraethylammonium cation. No other bands were observed at higher wavenumbers. The spectra are similar to those we reported earlier^{1,2} for the octahedral hexahalogenoprotactinates (IV) $[(\text{NEt}_4)_2\text{PaX}_6; \text{X} = \text{F}, \text{Cl} \text{ and } \text{Br}]$ and the optical absorption bands are similarly assigned to transitions from the Γ_7 ground level to the Γ_6, Γ_7' and Γ_8' states as indicated in Table 3.

The spectra are dominated by vibronic transitions which arise from the coupling of the vibrational modes with the pure electronic transitions. The strongest vibronic bands are those which are caused by motions which destroy the centre of symmetry of the complex during vibration [i.e. $\nu_3(\text{T}_{1u}), \nu_4(\text{T}_{1u})$ and $\nu_6(\text{T}_{2u})$]. In the PaI_6^{2-} electronic spectra it appears possible to correlate the vibronic band occurring at ca $\pm 145 \text{ cm}^{-1}$ from the $\Gamma_7 - \Gamma_7'$ transition with the $\nu_3(\text{T}_{1u})$ normal mode (Table 2). Other vibronic bands are probably due to other normal modes or combinations of normal modes including those of the cation¹¹. Higher resolution spectra are necessary to further resolve these bands. The Γ_8' level is again observed to be split as in the spectra of other octahedral protactinium (IV) complexes^{1,2} and a range of uranium (V) complexes^{12,13}.

The assignments at 85K shown in Table 3 were used to fit the ligand field splittings, θ and Δ , and the spin-orbit coupling constant³ and in order to have a check on these parameters the g value for PaI_6^{2-} was estimated at -1.15 (cf refs. 1 and 2). The experimental and calculated values are compared in Table 4, and the θ and Δ values are compared with those for other hexahalogenoprotactinate (IV) in Figure 4. The apparent increase in the spin-orbit coupling constant ζ through the series from the fluoride to the iodide complex should be treated with caution since the values obtained are strongly dependent on the g values estimated. Magnetic measurements are necessary on these compounds in order to determine accurate values of ζ .

The very marked decrease in Θ and the relatively constant value of Δ across the series $\text{PaF}_6^{2-} \rightarrow \text{PaI}_6^{2-}$ (Figure 4) reflects the progressive lowering in energy of the Γ_6 state relative to the Γ_8' and Γ_7' states shown in Table 5. The Γ_7' state in fact increases very slightly in energy from PaCl_6^{2-} to PaI_6^{2-} . As discussed previously, it can be shown for $5f'$ compounds in octahedral symmetry

that,

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$$\theta \approx (\alpha_{\pi}^2 + \alpha_{\sigma}^2 - \alpha_{\pi}'^2) (E_f - E_p)$$

and,

$$\Delta \approx (\alpha_{\pi}'^2) (E_f - E_p)$$

(1)

where α_{π} and α_{σ} are ligand admixture coefficients for the t_{1u} state, α_{π}' is that for the t_{2u} state, and E_f and E_p are the binding energies of the outer f metal ion electrons and the outer p shell ligand electrons, respectively. Thus, since θ changes much more rapidly than Δ for the hexahalogenoprotactinates(IV) (Figure 4) and α_{π} and α_{π}' involve the same ligand orbital, changes in π bonding will have relatively little effect on θ but (Eq.1) will directly effect Δ . It appears therefore that as the halide ion is changed from fluoride to iodide in the hexahalogenoprotactinates (IV), $(\text{NEt}_4)_2\text{PaX}_6$ ($X = \text{F}$ to I), large changes in σ bonding occur whilst the π bonding is relatively unaffected.

Summary

The preparation of actinide (IV) hexaiodo-complexes, $R_2^1\text{M}^{\text{IV}}\text{I}_6$ ($R^1 = \text{NEt}_4$, NMe_3 ; $\text{M}^{\text{IV}} = \text{Th}, \text{Pa}, \text{U}$) is described. I.r., Raman and electronic spectral data are reported for the protactinium (IV) and uranium (IV) complexes. Ligand field parameters and spin-orbit coupling constants derived for the hexaiodoprotactinates (IV) are discussed in relationship to earlier values obtained for other octahedral hexahalogenoprotactinates (IV), $(\text{NEt}_4)_2\text{PaX}_6$ ($X = \text{F}, \text{Cl}$ and Br) and shown to fit the trends previously identified for these compounds.

References.

1. N. Edelstein, D. Brown and B. Whittaker, *Inorg. Chem.*, 13, 563 (1974).
2. D. Brown, B. Whittaker and N. Edelstein, *Inorg. Chem.*, 13, 1805 (1974).
3. K.W. Bagnall, D. Brown, P.J. Jones and J.G.H. du Preez, *J. Chem. Soc.*, 350 (1965).
4. D. Brown and P.J. Jones, *J. Chem. Soc. (A)*, 243 (1967).
5. D. Brown and P.J. Jones, *J. Chem. Soc. (A)*, 247 (1967).
6. D. Brown, B. Whittaker and J. Edwards, Report AERE-R 7480 (1973).
7. D. Brown, B. Whittaker and P. Lidster, Report AERE-R 8035 (1975).
8. Bostik^{or} is a registered trade name. The material and appropriate thinner were purchased from Bostik Ltd., Leicester, England.
9. J. Shamir and A. Silberstein, *J. Inorg. Nucl. Chem.*, 32, 1173 (1975).
10. B.W. Berringer, J.B. Gruber, T.M. Loehr and G.P. O'Leary, *J. Chem. Phys.*, 55, 4608 (1971).
11. R.A. Satten, D. Young and D.M. Gruen, *J. Chem. Phys.*, 33, 1140 (1960).
12. D. Brown, B. Whittaker and N. Edelstein, Report AERE-R 7481 (1973).
13. J. Selbin and D. Ortego, *Chem. Rev.*, 69, 657 (1969).

Table 1

Partial X-ray Powder Diffraction Data for $(\text{NEt}_4)_2\text{I}_6$.

$\text{Sin}^2\theta_{\text{obs}}$	I^*_{est}	$\text{Sin}^2\theta_{\text{obs}}$	I^*_{est}
0.0108	M+	0.0716	S
.0115	M-	.0772	M-
.0293	W-	.0788	M-
.0331	W--	.0826	M
.0426	M	.0853	M
.0451	M	.0959	M+
.0464	W-	.0974	W-
.0501	W	.0996	W
.0511	W	.1045	M+
.0521	W-	.1080	W
.0545	W-	.1122	W+
.0563	W-	.1143	W-
.0571	W-	.1171	W-
.0604	W	.1261	M+
.0622	W-	.1327	M+
.0644	W	.1386	W
.0666	W-	.1421	W
* .0689	W-	.1471	W
		.1542	W-

* S, strong; M, medium; W, weak.

Table 2.

I.r. and Raman Data For Hexaiodo-complexes.

<u>Compound</u>	<u>Colour</u>	<u>Observed Vibrational Modes (cm^{-1})</u>			
		ν_1	ν_3	ν_4	ν_5
$(\text{NEt}_4)_2\text{ThI}_6$	White	131	143	<u>c</u>	$68,50^{\text{c}}$
$(\text{NEt}_4)_2\text{PaI}_6$	Blue	<u>a</u>	143	<u>c</u>	<u>a</u>
$(\text{NEt}_4)_2\text{UI}_6$	Red	<u>b</u>	143	<u>c</u>	<u>b</u>

a not observed owing to fluorescence

b not observed owing to rapid decomposition of compound in laser beam

c discussed in the text.

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Table 3. Spectral Results For The Hexaiodoprotactinates (IV)^a

At 85K			<u>(NEt₄)₂PaI₆</u>			At 300K			<u>(NMe₃Ph)₂PaI₆</u>		
Band Position (cm ⁻¹)	Int.	Freq. (cm ⁻¹)	Band Position (cm ⁻¹)	Int.	Freq. (cm ⁻¹)	Band Position (cm ⁻¹)	Int.	Freq. (cm ⁻¹)	Band Position (cm ⁻¹)	Int.	Freq. (cm ⁻¹)
Γ ₇ -Γ ₆	w	+298	7262	sh	+298	7413	w	+420			
	s	+174	7138	s	+179	7315	m	+322			
	sh	+138	7102			7143	s	+150	7143	s, br	+145
	m	0	6964	w	0	6993	m	0	6998	w	0
	m	-96	6868	s	-105	6887	sh	-106			
	m	-194	6770	s	-184	6840	s	-153	6840	s, br	-158
sh	-235	6729			6752	w	-241				
Γ ₇ -Γ ₈ '	s		6601	s		6627	s		6614	s, br	
	s		6502	s		6494	s		6398	s, br	
Γ ₇ -Γ ₇ '	w	+152	5534	m	+140	5540	w	+155	5540	m	+149
	w	+50	5432	m	+44	5423	w	+38	5429	m	+38
	sh	0	5382	m	0	5385	m	0	5391	m	0
	m	-23	5359	m	-23	5362	m	-23	5371	m	-20
				sh	-35	5333	sh	-107	5330	sh	-61
	w, b	-141	5241	w	-141	5236	w, br	-149	5241	w	-150

^a s, strong; m, medium; w, weak; br, broad; sh, shoulder.

Table 4.

Calculated and Experimental Transition Energies, Ligand Field Parameters
and Spin-Orbit Coupling Constants For Hexaiodoprotactinates (IV).

<u>Compound</u>		<u>Energy (cm⁻¹)</u>			<u>Parameters (cm⁻¹)</u>			
		$\Gamma_7-\Gamma_6$	$\Gamma_7-\Gamma_8'$	$\Gamma_7-\Gamma_7'$	ξ	θ	Δ	ζ
$(\text{NEt}_4)_2\text{PaI}_6$	Calc.	7002	6497	5397	-1.142	832	1546	1542
	Exp.	6998	6506	5391	-1.150 ^a			
$(\text{NMe}_3\text{Ph})_2\text{PaI}_6$	Calc.	6973	6497	5399	-1.130	782	1592	1540
	Exp.	6964	6518	5385	-1.150 ^a			

^a Estimated value.

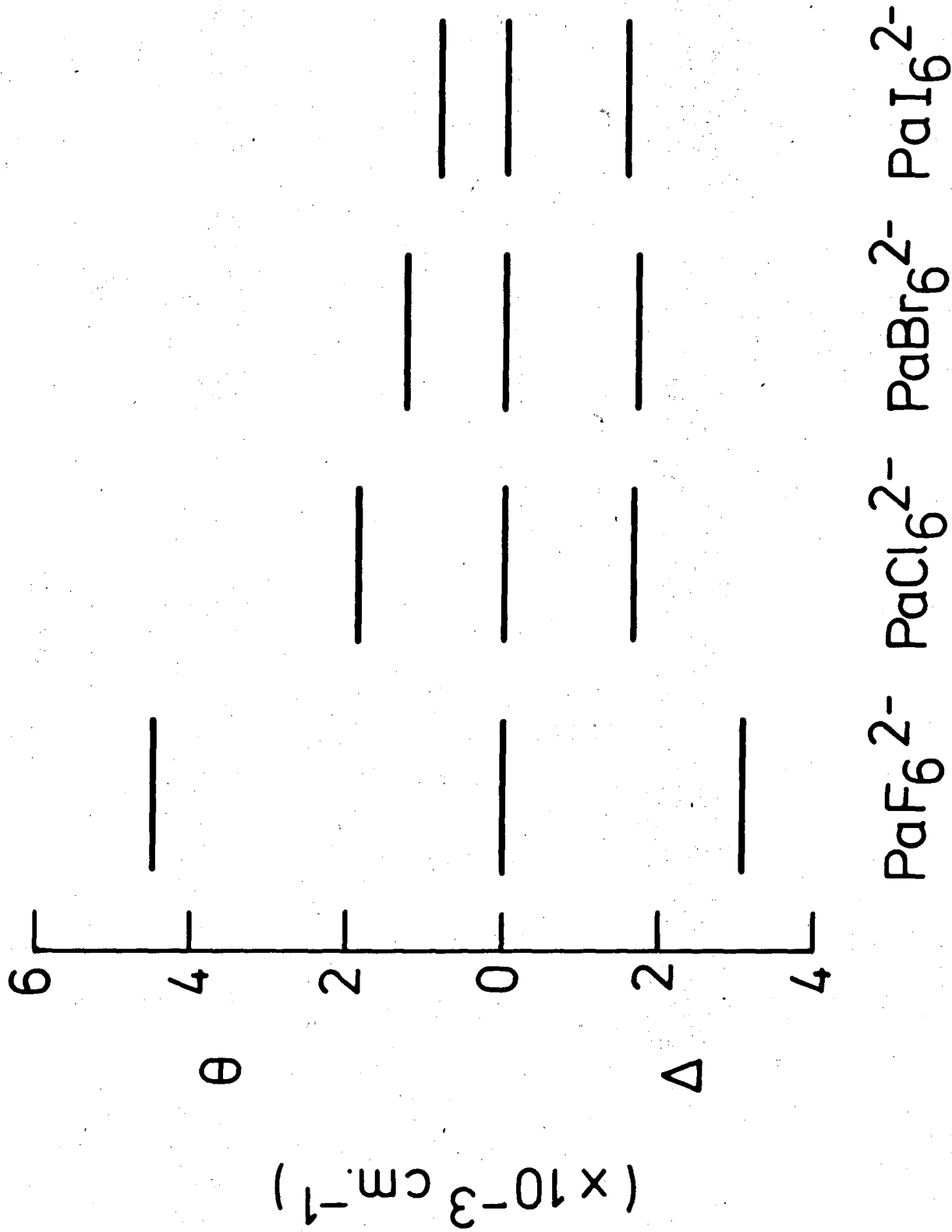
Table 5.Comparison of Experimental Transition Energies for Hexahalogenoprotactinates (IV).

<u>Compound</u>		<u>Energy (cm⁻¹)</u>		
		$\Gamma_7-\Gamma_6$	$\Gamma_7-\Gamma_{8'}$	$\Gamma_7-\Gamma_{7'}$
$\alpha-(\text{NEt}_4)_2\text{PaF}_6$	<u>a</u>	11,446	9,708	5,698
$(\text{NEt}_4)_2\text{PaCl}_6$	<u>b</u>	8,011	7,022	5,330
$(\text{NEt}_4)_2\text{PaBr}_6$	<u>b</u>	7,480	6,828	5,365
$(\text{NEt}_4)_2\text{PaI}_6$	<u>c</u>	6,998	6,506	5,391
$(\text{NMe}_3\text{Ph})_2\text{PaI}_6$	<u>c</u>	6,964	6,518	5,385

a Ref 2; b Ref 1; c This work.

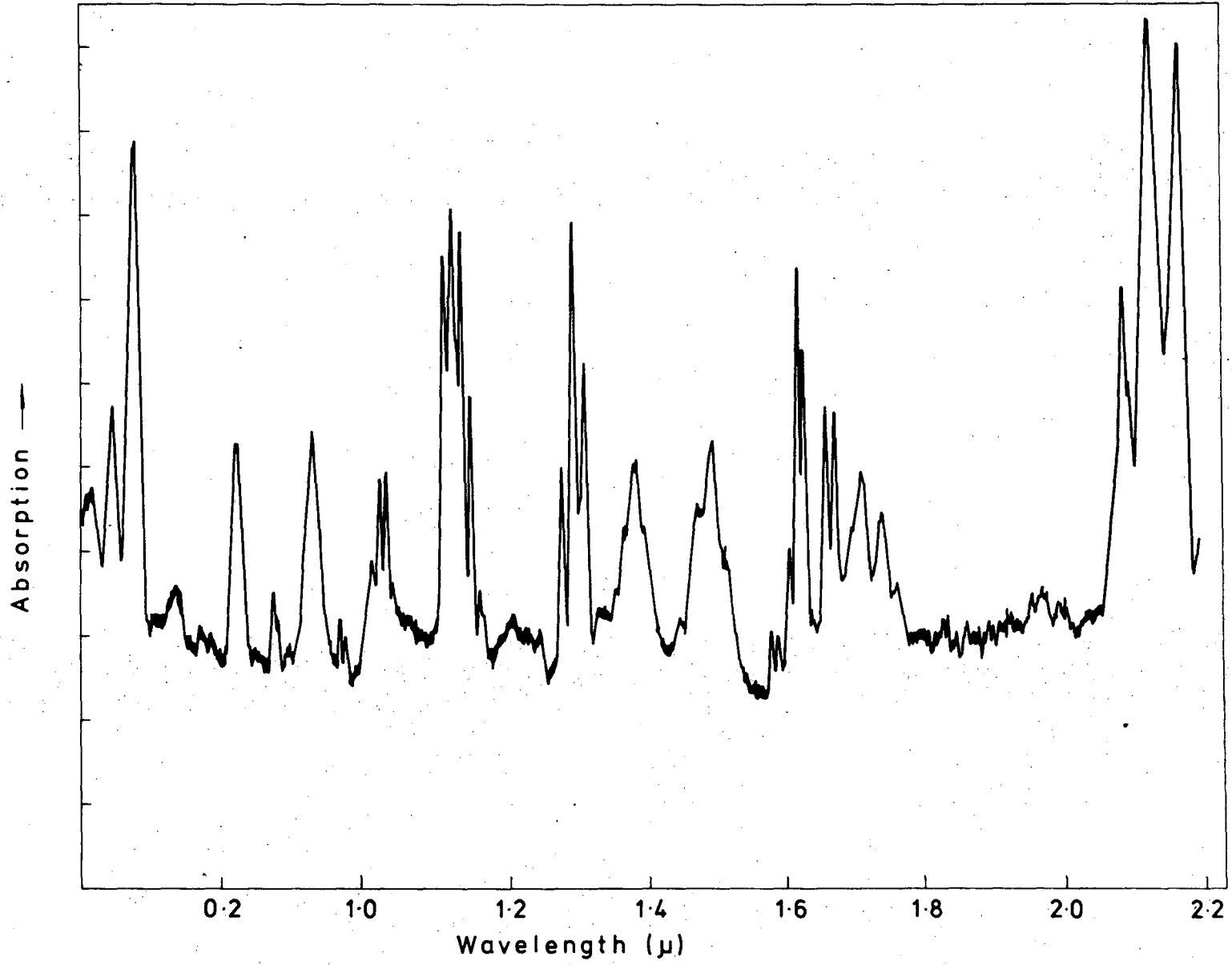
Figure Captions.

- Figure 1 The Spectrum of $(\text{NEt}_4)_2\text{PaI}_6$ at 300K
- Figure 2 The Spectrum of $(\text{NEt}_4)_2\text{PaI}_6$ at 85K
- Figure 3 The Spectrum of $(\text{NEt}_4)_2\text{UI}_6$ at 85K
- Figure 4 Ligand Field Splittings θ and Δ for octahedral
Complexes of the type $(\text{NEt}_4)_2\text{PaX}_6$ (X=F, Cl, Br and I).



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Fig. 1



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Fig. 2

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LBL-4083

STATISTICAL DECAY OF GAMMA RAYS IN (n_{th}, γ) REACTIONS

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(Revised 3 September 1974)

Abstract: Isomer ratios obtained in doubly even targets in the (n_{th}, γ) reaction are calculated using realistic level densities and spin cut-off parameters. The calculations were performed using the Monte Carlo method and the general agreement between the present calculations and experimental data is satisfactory. In one case the shape of the γ -ray spectrum was calculated and good agreement with experimental data was obtained.

1. Introduction

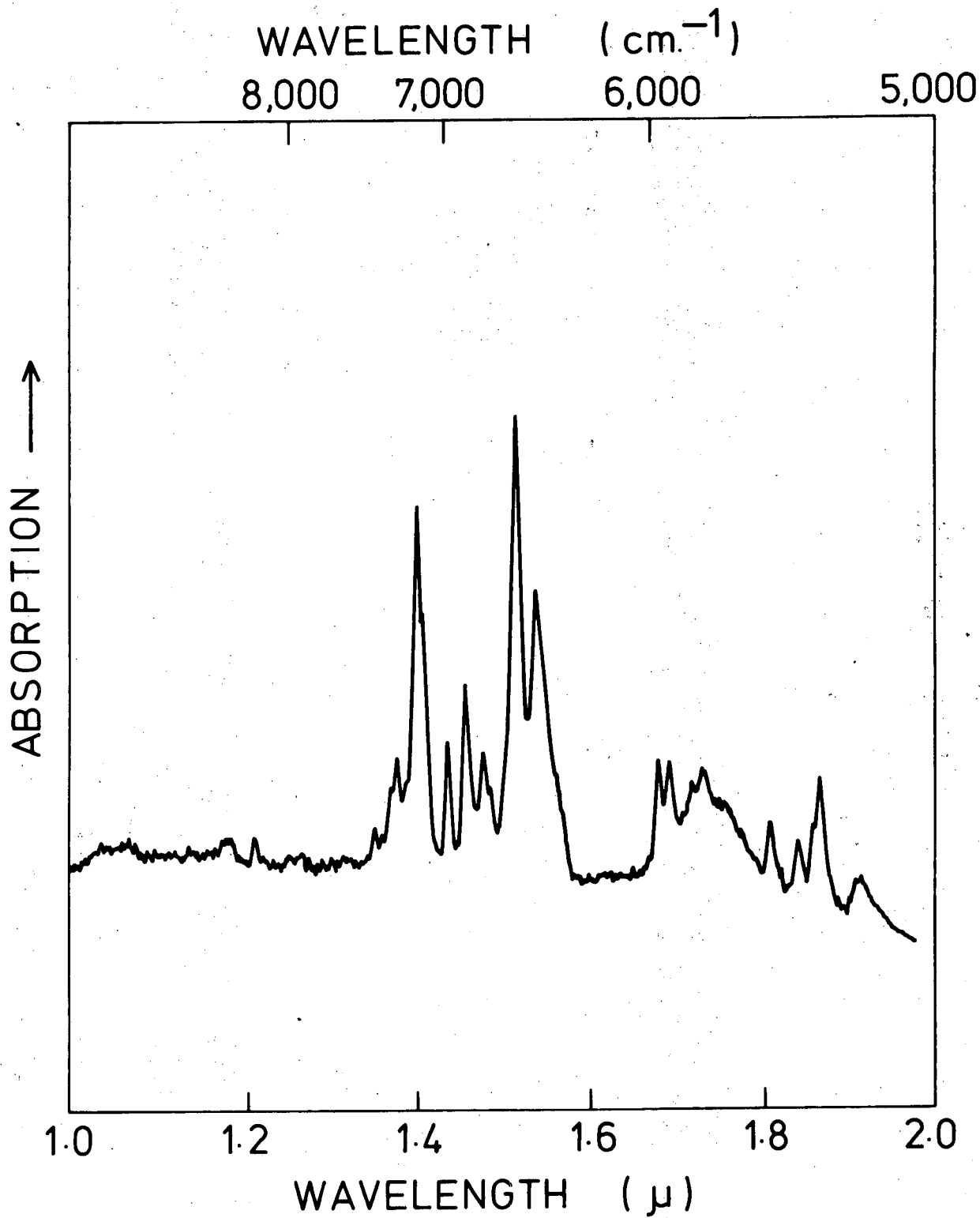
The relative formation probability of a spin isomer and of the ground state following a nuclear reaction has been studied extensively in many types of nuclear reactions. The isomer ratio which is obtained from these reactions is defined as the ratio of the yield of the high spin isomer to the total yield of the ground and isomer state. In the present paper we calculate isomer ratios in (n_{th}, γ) reactions for a large number of doubly even target nuclei on the basis of the statistical model of the nucleus. We shall also calculate in one case the shape of the statistical portion of the gamma ray spectrum using the same theoretical model.

The first quantitative calculation of spin isomer formation was carried out by Vandenbosch and Huizenga^{1, 2)} and later by Vonach, Vandenbosch and Huizenga³⁾ and Bishop *et al.*⁴⁾. These authors assumed that the gamma cascade in the (n_{th}, γ) reaction is composed of E1 transitions only, the number and energy of which were obtained on the basis of the excitation energy and the value of a , the level density parameter^{3, 4)}.

The isomer ratio calculations were considerably improved by Sperber and co-workers^{5, 6)}. In particular the number of transitions in the γ -cascade was allowed to vary and the isomeric and ground states were also fed during the gamma cascade and not only in the last step of the deexcitation process. The level density and spin cut-off parameters were taken from Gilbert and Cameron⁷⁾ while the degree of quadrupole admixture was a free parameter.

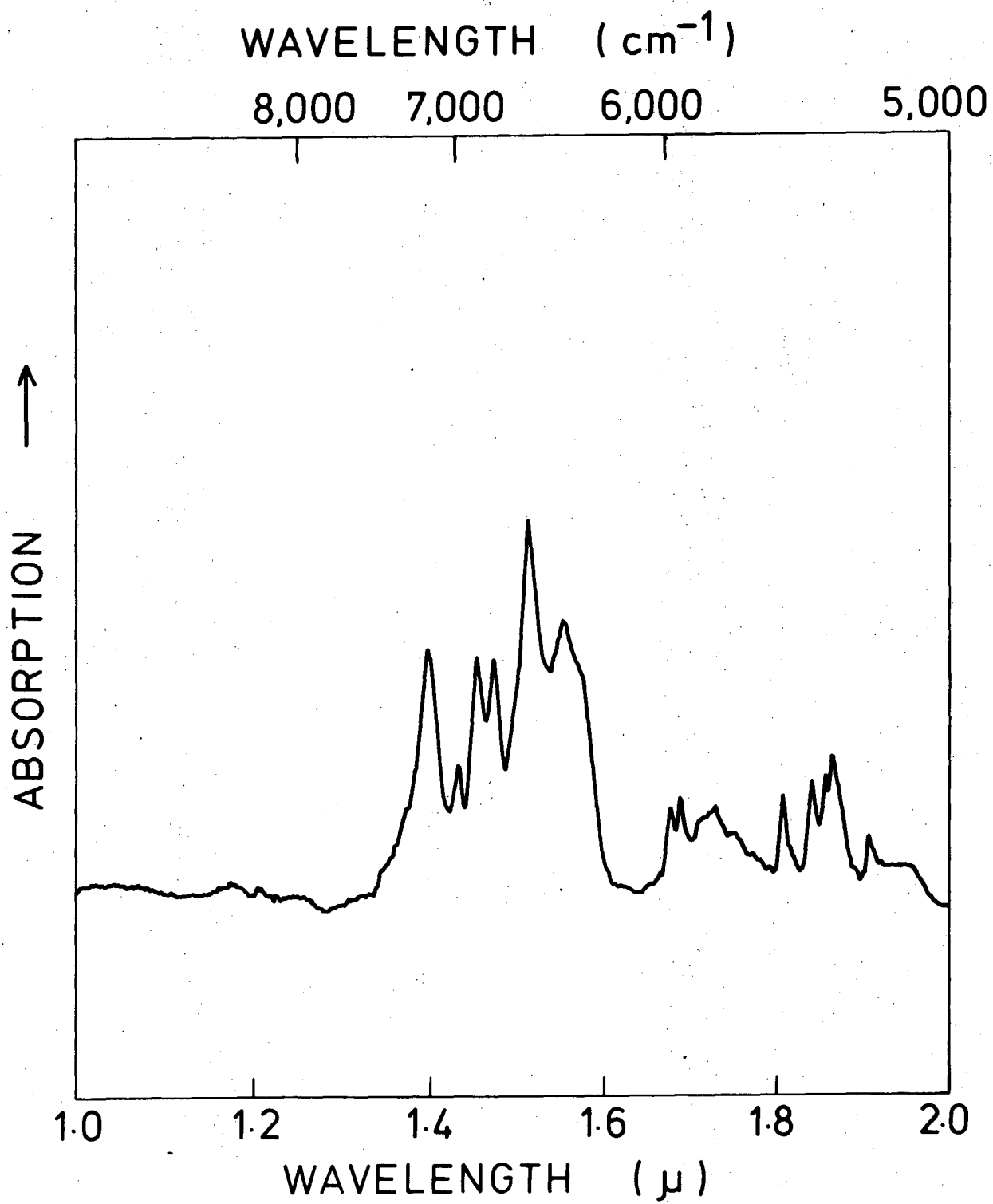
Recently, Watson *et al.*⁸⁾ and Clarke and Gill⁹⁾ used the shell-model based

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XBL 758-1890

Fig. 3



XBL 758-1891

Fig. 4

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