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Calculated Photoemission Spectra of the 5f States in the Actinides

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Abstract

The complex multiplet structure of the partially filled 5f shell in the Actinides is calculated in intermediate coupling. The obtained eigenfunctions are used for a calculation of photoemission intensities in the fractional parentage scheme including all final states. The obtained calculated spectra are significantly different from the equivalent results for the 4f states in the Rare Earth metals presented recently.

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The investigation of several actinide-metals and -compounds in photoemission spectroscopy increased during the last years (Veal et al. 1977, Naegele et al. 1980, Naegele et al. 1982, Naegele et al. 1983). Although the 5f-electrons in the actinide-metals are not as localized as the 4f-electrons in the rare earth-metals a multiplet structure is expected in 5f-photoemission spectra of some actinide-compounds and -oxyds and for actinide metals with more than 5 electrons in the 5f-shell (Smith et al. 1983). For the rare earth-metals a calculation in the fractional parentage scheme of the expected photoemission intensities of the 4f--multiplet structure has been presented previously (Gerken 1983) and showed an excellent agreement with X-ray photoemission experiments (Lang et al. 1981). This calculation proved to be also extremely useful for the interpretation of synchrotron radiation spectra of the rare earths taken at HASYLAB/DESY (Gerken 1982).

The first metal in the series of the actinides for which a 5f-multiplet structure is expected in photoemission spectra is Am with six 5f-electrons in the ground state. The measured spectra (Naegele et al. 1982, 1983) show completely different structures compared to the corresponding $4f^6 \rightarrow 4f^5$ emission of the Sm surface (Gerken 1982, Gerken et al. 1982). This result had to be expected since the ratio of the Coulomb-interaction and spin-orbital interaction is different in the rare earths and in the actinides.

Therefore we present in this paper a complete calculation in intermediate coupling for the energy positions of the $5f^N$ multiplet lines and expected photoemission intensities for all possible $5f^N \rightarrow 5f^{N-1}$ transitions (N = 2 - 13) analogous to the calculation for the rare earth metals (Gerken 1983). The result is tabulated and the multiplet lines convoluted with the lineshapes of Doniach and Sunjic (1970) are presented as calculated spectra.Lifetime width and asymmetry were chosen identical to those taken for the calculated rare earth metal spectra to provide a simple estimate of the differences between both calculated results.

A general outline of the theory is given by Gerken (1983). Therefore only the results will be presented here.

The parameter used for the energy level calculation are comprised in table 1. They were taken from different publications (see table 1, last column) in which absorption spectra of actinide ions were fitted with calculated lines in intermediate coupling. In some of their calculations the authors used several parameters to account for additional magnetic interactions

which can be treated in a third order perturbation theory and for crystal field splitting in actinide-halide samples which have been investigated. A comparison of two sets of parameter for Pu^{3+} with (Crosswhite et al. 1980) and without additional parameters (Carnall et al. 1970) showed only a difference of about 5 % in the Coulomb interaction parameters E^1, E^2 and E^3 while the spin-orbit interaction parameter ξ_{5f} remained nearly unchanged. Therefore for simplicity in our calculation this additional parameters were omitted.

For the configuration $5f^2$, $5f^{12}$ and $5f^{13}$ we could not find published parameters and therefore these values were extrapolated from fig. 1 and fig. 2 in which the parameters for E^1 and ξ_{nf} are shown for all 4f- and 5f-configurations. E^2 and E^3 are proportional to E^1. The configuration interaction parameters α, β and γ show only small variations for the different configurations and consequently we used values similar to the neighbouring elements for the extrapolated parameters. From this figures it can easily be seen that the parameters for the 4f- and 5f-configurations show very similar variations. As already pointed out by Carnall et al. (1968) the electrostatic interaction parameters can be fitted by a linear function while the best fit for the spin-orbital interaction parameters are two linear functions with different gradients for the less than half filled and more than half filled shells. Fig. 1 and fig. 2 also demonstrate the drastic change in the importance of the Coulomb- and spin-orbital-interaction between 4f- and 5f-configurations. This results in an even stronger breakdown of the LS-coupling approximation in the actinides than in the rare earths.

The total energy matrices were diagonalized for all states from the configurations $5f^2$ to $5f^{13}$. The resultant eigenvalues represent the energy levels of the different states of a configuration and the pertinant eigenfunctions give the composition of this intermediate coupling states in LS-basis. The lowest energy level of each $5f^N$-configuration represents its ground state and was therefore used as initial state in the intensity calculation which was performed for all $5f^N \rightarrow 5f^{N-1}$ (N = 2 - 13) transitions. The results are given in table 2. The different final states are characterized by the main LS-contribution of the eigenfunction while the initial states are given completely. Even the ground state of the different $5f^N$ configurations show a strong mixing of several LS-levels and demonstrate the degree of the breakdown of the LS-coupling approximation. This fact is

confirmed by table 3 which gives the eigenfunctions of the f^2 ground-state (J = 4) for the LS-coupling-limit, the jj-coupling limit and in intermediate coupling for $4f^2$ in Pr^{3+} and $5f^2$ in Pa^{3+} as well as the corresponding photoemission intensities for the $f^2 \rightarrow f^1$ transition. It is obvious that the $5f^2$-configuration in Pa^{3+} is much closer to the jj-limit than the $4f^2$-configuration in Pr^{3+}. This results are in good agreement with similar calculations with jj-coupling basis states by Beatham et al. (1979).

In order to illustrate our calculated results from table 2, the multiplet lines are convoluted with the lineshapes of Doniach and Sunjic (1970) and with Gaussian profiles to account for a hypothetical experimental resolution, and then plotted on a binding energy scale relative to the lowest multiplet line (figure 3). The parameters are listed in table 4. The spectra are scaled in energy by a factor of 1.1 compared with the energy positions listed in table 2 in order to account for the different nuclear charge in photoemission final states and the absorption experiments.

Compared to the calculated spectra for the rare earth metals (Gerken 1983) strong differences are obvious. The multiplet splitting is somewhat reduced in the $5f^N$-configurations compared to the $4f^N$-configurations and the photoemission intensities in the $5f^N$ configurations are shifted to the lines with relative low binding energies.

Our results disagree completely with a similar work by Veal et al. (1977). The authors calculated the photoemission spectra for the transition $5f^N \rightarrow 5f^{N-1}$ for N = 3 - 8 using a formulae which must be strongly questioned because it sums up intensities and ignores possible interference effects by adding up amplitudes of the different intermediate coupling states (compare Gerken 1983 equ. (11) and Veal et al. 1977 equ. (A4), see also Cox et al. 1981 equ. (10)).

There is, however, no agreement between our calculated spectrum and the measured $5f^6 \rightarrow 5f^5$ multiplet structure in Am metal (Naegele et al. 1982, 1983) which is the first metal for which localized 5f electrons are expected. Unfortunately high resolution spectra are only measured for relative low photon energies (20 to 50 eV) where the surface sensitivity is extremely

high and surface shifts as well as valence changes at the surface as observed in the rare earth metals (Kammerer et al. 1982, Gerken 1982, Gerken et al. 1982, 1983) may complicate the spectra.

For a test of the calculated spectra additional experiments on other $5f^N$-systems with less surface sensitivity are indispensable.

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Figure Captions

Table 1: Radial energy parameters which were used to diagonalize the total energy matrices. Energies are given in cm^{-1}. The values are taken from the references cited in the last column. The extrapolated values are taken from Fig. 1 and Fig. 2.

Table 2: Energy positions and intensities of the mulitplet lines for the transitions $5f^N \rightarrow 5f^{N-1}$ with N = 3 ~ 13. Intensities lower than 0.01 are omitted. For the initial states the complete eigenfunctions and for the final state only the main contribution (absolute value) in the representation $(w_1\ w_2\ w_3)$, $(u_1\ u_2)$, seniority, $2S + 1$, L are given.

Table 3: Calculated photoemission intensities for the transition $f^2 \rightarrow f^1$ in the LS-coupling limit, the jj-coupling limit and in intermediate coupling ($4f^2 \rightarrow 4f^1$ in Pr^{3+} and $5f^2 \rightarrow 5f^1$ in Pa^{3+}). The initial state eigenfunctions are given in LS-basis states.

Table 4: Parameters of the curves of Doniach and Sunjic (1970) from figure 3.

Fig. 1: Energy values of the radial parameter E^1 for the different $4f^N$ (see Gerken 1983) and $5f^N$-configurations (see table 1).

Fig. 2: Energy values of the spin-orbital interaction parameter ξ for the different $4f^N$ (see Gerken 1983) and $5f^N$-configurations (see table 1).

Fig. 3: Calculated spectra of the different $5f^N \rightarrow 5f^{N-1}$ transitions in the actinides on a binding energy scale relative to the lowest mulitplet line. The calculated multiplet lines from table 2 are convoluted with the lineshapes of Doniach and Sunjic (1970) using the parameters listed in table 4. A calculated background (Gerken 1982) is added and the resultant curve folded with a Gaussian profile of 0.2 eV FWHM to account for an hypothetical experimental resolution.

Table 1

f^N	Element	E^1	E^2	E^3	ξ_{5f}	α	β	γ	Parameter reference
f^2	Pa^{3+}	2650	11.3	245.4	1350	28	-730	1100	extrapolated
f^3	U^{3+}	2878.5	11.848	258.25	1623.0	27.6	-722	1000	Crosswhite et al. 1980
f^4	Np^{3+}	3168.9	13.679	297.75	1932.6	32.713	-756.12	1084.2	Carnall et al. 1974
f^5	Pu^{3+}	3634.5	15.356	342.15	2272.2	31.064	-675.0	29.743	Carnall et al. 1970
f^6	Am^{3+}	3582.8	17.276	334.30	2593.3	21.634	-158.48	1240.4	Pappalardo et al. 1969
f^7	Cm^{3+}	3955.4	17.789	369.79	2876.1	27.895	-925.21	1119.6	Carnall et al. 1975
f^8	Bk^{3+}	4127.2	19.537	387.83	3252.8	31.123	-1283.7	1247.5	Carnall et al. 1973a
f^9	Cf^{3+}	4173.2	19.84	391.40	3601.7	35.371	-748.02	1200.0	Carnall et al. 1973b
f^{10}	Es^{3+}	4445.9	21.446	418.64	4014.7	22.505	-722.53	1000.0	Carnall et al. 1973c
f^{11}	Fm^{3+}	4800	22.9	453	4450	23	-730	1100	extrapolated
f^{12}	Md^{3+}	5000	23.8	472	4900	23	-730	1100	extrapolated

Photoemission-Intensity in intermediate coupling
Transition from 5f 3 to 5f 2

Initial state eigenfunction: J = 4.5

0.0277	(111)(10)	3	4	F
0.0487	(111)(20)	3	4	G
0.9155	(111)(20)	3	4	I
-0.0793	(210)(20)	3	2	G
0.0774	(210)(21)	3	2	G
0.1099	(210)(11)	3	2	H
-0.3666	(210)(21)	3	2	H

Final state intensities:

					J	E (eV)	Intensity
0.93	(110)(10)	2	3	F	2.0	0.3687	0.6209
1.00	(110)(10)	2	3	F	3.0	0.7726	0.0314
0.95	(110)(11)	2	3	H	4.0	0.0000	2.2134
1.00	(110)(11)	2	3	H	5.0	0.5160	0.1274

Photoemission-Intensity in intermediate coupling
Transition from 5f 4 to 5f 3

Initial state eigenfunction: J = 4.0

-0.0409	(111)(20)	4	5	G
0.8985	(111)(20)	4	5	I
0.0318	(211)(30)	4	3	F
0.0862	(211)(21)	4	3	G
-0.0575	(211)(30)	4	3	G
0.1918	(110)(11)	2	3	H
0.1901	(211)(21)	4	3	H
-0.3051	(211)(30)	4	3	H
0.0518	(200)(20)	2	1	G
0.0852	(220)(22)	4	1	G

Final state intensities:

					J	E (eV)	Intensity
0.80	(111)(0)	3	4	S	1.5	1.2725	0.0273
0.80	(111)(10)	3	4	F	1.5	0.7828	0.3532
0.80	(111)(20)	3	4	G	2.5	1.2711	0.3171
0.78	(111)(10)	3	4	F	2.5	1.1203	0.5706
0.85	(111)(20)	3	4	G	3.5	1.5178	0.1261
0.92	(111)(20)	3	4	I	4.5	0.0000	2.2604
0.98	(111)(20)	3	4	I	5.5	0.5141	0.2806
0.97	(111)(20)	3	4	I	6.5	0.9616	0.0107

Photoemission-Intensity in intermediate coupling
Transition from 5f 5 to 5f 4

Initial state eigenfunction: J = 2.5

0.1075	(110)(10)	5	6	F
0.8124	(110)(11)	5	6	H
-0.1285	(211)(21)	5	4	F
-0.3145	(111)(20)	3	4	G
0.1218	(211)(21)	5	4	G
-0.3700	(211)(30)	5	4	G
-0.0693	(100)(10)	1	2	F
-0.0951	(210)(21)	3	2	F
-0.1034	(221)(31)	5	2	F
-0.1014	(221)(31)	5	2	F

Final state intensities:

					J	E (eV)	Intensity
0.83	(111)(20)	4	5	D	0.0	2.1881	0.1984
0.73	(111)(20)	4	5	D	1.0	2.4955	0.0992
0.58	(111)(20)	4	5	D	1.0	2.3911	0.0569
0.90	(111)(10)	4	5	F	1.0	1.1218	0.1550
0.85	(111)(20)	4	5	D	2.0	2.7661	0.0254
0.48	(111)(10)	4	5	F	2.0	2.1899	0.0195
0.78	(111)(0)	4	5	S	2.0	1.4975	0.0104
0.75	(111)(20)	4	5	G	2.0	1.3929	0.1599
0.59	(111)(10)	4	5	F	2.0	0.9191	0.8194
0.63	(211)(21)	4	3	G	3.0	2.2228	0.0147
0.81	(111)(20)	4	5	G	3.0	1.4906	0.3981
0.85	(111)(10)	4	5	F	3.0	1.3471	0.0513
0.76	(111)(20)	4	5	G	4.0	2.1126	0.0296
0.80	(111)(10)	4	5	F	4.0	1.7977	0.0185
0.40	(211)(21)	4	3	G	4.0	1.2686	0.0170
0.90	(111)(20)	4	5	I	4.0	0.0000	2.2660
0.95	(111)(20)	4	5	I	5.0	0.4583	0.4283

Photoemission-Intensity in intermediate coupling
Transition from 5f 6 to 5f 5

Initial state eigenfunction: J = 0.0

0.6704	(100)(10)	6	7	F
-0.4438	(111)(20)	4	5	D
0.4290	(210)(21)	6	5	D
-0.1882	(110)(11)	2	3	F
0.2022	(211)(30)	4	3	F
0.0410	(221)(30)	6	3	F
0.2613	(221)(31)	6	3	F
-0.0456	(0)(0)	0	1	S
-0.1035	(220)(22)	4	1	S
0.0950	(222)(40)	6	1	S

Final state intensities:

					J	E (eV)	Intensity
0.61	(110)(11)	5	6	F	2.5	2.9621	0.0635
0.54	(110)(11)	5	6	F	2.5	2.1183	0.2670
0.59	(110)(10)	5	6	F	2.5	1.7330	0.0287
0.70	(110)(10)	5	6	F	2.5	0.8278	1.3253
0.81	(110)(11)	5	6	H	2.5	0.0000	3.5180
0.82	(110)(10)	5	6	F	3.5	1.1710	0.0769
0.90	(110)(11)	5	6	H	3.5	0.4140	0.6691

Photoemission-Intensity in intermediate coupling
Transition from 5f 7 to 5f 6

Initial state eigenfunction: J = 3.5

0.8931	(0)(0)	7	8	S
0.4158	(110)(11)	5	6	F
-0.0894	(200)(20)	7	6	D
0.0214	(110)(10)	5	6	F
0.0966	(111)(20)	3	4	D
0.0264	(211)(21)	5	4	D
-0.0181	(220)(20)	7	4	D
0.0927	(220)(22)	7	4	D
-0.0307	(211)(30)	5	4	F
-0.0120	(100)(10)	1	2	F
-0.0190	(221)(31)	5	2	F
-0.0104	(221)(31)	5	2	F

Final state intensities:

				J	E (eV)	Intensity
0.67	(100)(10)	6	7 F	0.0	1.4988	0.0216
0.67	(100)(10)	6	7 F	0.0	0.0000	0.1651
0.55	(210)(21)	6	5 D	1.0	2.1706	0.0159
0.81	(100)(10)	6	7 F	1.0	0.3632	0.5031
0.88	(100)(10)	6	7 F	2.0	0.6757	0.7736
0.91	(100)(10)	6	7 F	3.0	0.9478	1.0126
0.91	(100)(10)	6	7 F	4.0	1.2020	1.2433
0.47	(210)(21)	6	5 H	5.0	2.9083	0.0103
0.89	(100)(10)	6	7 F	5.0	1.4145	1.4782
0.42	(210)(21)	6	5 L	6.0	3.0575	0.0232
0.62	(210)(21)	6	5 L	6.0	2.3826	0.0231
0.84	(100)(10)	6	7 F	6.0	1.5933	1.6750

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Photoemission-Intensity in intermediate coupling
Transition from 5f 8 to 5f 7

Initial state eigenfunction: J = 6.0

0.8848	(100)(10)	6	7	F
-0.3188	(111)(20)	4	5	G
0.0925	(210)(20)	6	5	G
0.2936	(210)(21)	6	5	G
0.0403	(210)(11)	6	5	H
0.0681	(210)(21)	6	5	H
0.0197	(210)(20)	6	5	I
-0.0507	(110)(11)	2	3	H
0.0629	(211)(21)	4	3	H
0.0531	(211)(30)	4	3	H
0.0229	(221)(30)	6	3	H
0.0256	(221)(31)	6	3	H
0.0429	(221)(31)	6	3	H
-0.0162	(211)(30)	4	3	I
-0.0168	(221)(31)	6	3	I

Final state intensities:

				J	E (eV)	Intensity
0.67	(110)(11)	5	6 P	2.5	2.5720	0.2519
0.47	(110)(10)	5	6 F	3.5	4.6819	0.0152
0.51	(110)(11)	5	6 P	3.5	3.6461	0.0454
0.76	(200)(20)	7	6 I	3.5	2.8307	0.0190
0.49	(200)(20)	7	6 D	3.5	2.3120	0.6202
0.89	(0)(0)	7	8 S	3.5	0.0000	1.2925
0.47	(220)(21)	7	4 H	4.5	5.1883	0.0193
0.44	(110)(10)	5	6 F	4.5	4.7527	0.0105
0.59	(200)(20)	7	6 G	4.5	4.0469	0.0510
0.62	(200)(20)	7	6 D	4.5	3.2245	0.4048
0.66	(200)(20)	7	6 I	4.5	2.9496	0.3316
0.48	(110)(10)	5	6 F	5.5	4.3769	0.0187
0.69	(200)(20)	7	6 G	5.5	3.9063	0.8449
0.84	(200)(20)	7	6 I	5.5	3.1550	0.1319
0.76	(200)(20)	7	6 G	6.5	4.3149	0.8632
0.81	(200)(20)	7	6 I	6.5	3.2069	0.2266
0.47	(220)(22)	7	4 I	7.5	6.8505	0.0173
0.43	(220)(20)	7	4 I	7.5	5.9407	0.0298
0.58	(220)(21)	7	4 L	7.5	5.2009	0.0547
0.72	(110)(11)	5	6 H	7.5	4.9379	0.5590
0.86	(200)(20)	7	6 I	7.5	3.2846	0.6197
0.46	(220)(22)	7	4 N	8.5	4.3157	0.0440
0.85	(200)(20)	7	6 I	8.5	3.1398	1.3645

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Photoemission-Intensity in intermediate couplins
Transition from 5f 9 to 5f 8

Initial state eisenfunction: J = 7.5

0.8682	(110)(11)	5	6	H
-0.2253	(111)(20)	3	4	I
-0.4118	(211)(30)	5	4	I
-0.0929	(211)(21)	5	4	K
0.0375	(211)(30)	5	4	K
0.0372	(211)(21)	5	4	L
0.0791	(210)(21)	3	2	K
-0.0309	(221)(30)	5	2	K
0.0578	(221)(31)	5	2	K
-0.0389	(221)(31)	5	2	K

Final state intensities:

					J	E (eV)	Intensity
0.49	(210)(21)	6	5	F	4.0	3.1554	0.0255
0.40	(210)(11)	6	5	H	4.0	2.4029	0.0241
0.70	(100)(10)	6	7	F	4.0	1.7797	0.0885
0.65	(100)(10)	6	7	F	4.0	0.3665	0.5053
0.49	(210)(20)	6	5	G	5.0	3.7510	0.0103
0.54	(210)(21)	6	5	K	5.0	3.6535	0.0120
0.46	(210)(21)	6	5	F	5.0	2.9173	0.1809
0.47	(210)(11)	6	5	H	5.0	2.6641	0.0568
0.91	(100)(10)	6	7	F	5.0	0.5585	0.5776
0.45	(210)(21)	6	5	H	6.0	4.4556	0.0144
0.57	(210)(21)	6	5	K	6.0	4.0123	0.0290
0.53	(210)(11)	6	5	H	6.0	3.6425	0.2189
0.44	(210)(21)	6	5	H	6.0	2.7669	0.3939
0.46	(210)(21)	6	5	L	6.0	1.9253	0.1767
0.88	(100)(10)	6	7	F	6.0	0.0000	1.8418
0.58	(111)(20)	4	5	I	7.0	6.3743	0.0107
0.57	(210)(21)	6	5	K	7.0	4.7943	0.0404
0.61	(210)(21)	6	5	H	7.0	4.4119	0.1484
0.63	(210)(20)	6	5	I	7.0	4.2383	0.0374
0.51	(210)(11)	6	5	H	7.0	3.5632	0.0255
0.62	(210)(21)	6	5	L	7.0	3.2256	0.1877
0.43	(210)(21)	6	5	L	7.0	2.3299	0.4906
0.53	(111)(20)	4	5	I	8.0	6.2557	0.0803
0.67	(111)(20)	4	5	I	8.0	6.0801	0.1307
0.43	(221)(30)	6	3	M	8.0	5.7708	0.0211
0.40	(221)(21)	6	3	K	8.0	5.2023	0.0146
0.76	(210)(21)	6	5	K	8.0	4.0938	0.1251
0.71	(210)(20)	6	5	I	8.0	3.6176	0.1401
0.63	(210)(21)	6	5	L	8.0	2.7791	0.5488
0.56	(221)(31)	6	3	N	9.0	4.3877	0.0854
0.67	(210)(21)	6	5	K	9.0	3.8222	0.2869
0.73	(210)(21)	6	5	L	9.0	2.7801	0.8227
0.53	(221)(31)	6	3	N	10.0	4.0754	0.0484
0.82	(210)(21)	6	5	L	10.0	2.5251	1.2703

Photoemission-Intensity in intermediate couplins
Transition from 5f10 to 5f 9

Initial state eisenfunction: J = 8.0

0.8650	(111)(20)	4	5	I
0.2244	(211)(21)	4	3	K
-0.4140	(211)(30)	4	3	K
-0.1109	(211)(21)	4	3	L
-0.0382	(211)(30)	4	3	M
0.0429	(220)(21)	4	1	L
0.1204	(220)(22)	4	1	L

Final state intensities:

					J	E (eV)	Intensity
0.59	(110)(11)	5	6	H	4.5	2.6189	0.0284
0.53	(110)(10)	5	6	F	4.5	0.7053	0.5150
0.43	(211)(20)	5	4	G	5.5	3.9331	0.0174
0.46	(211)(30)	5	4	I	5.5	2.8936	0.0285
0.69	(110)(11)	5	6	H	5.5	2.4173	0.1105
0.69	(110)(10)	5	6	F	5.5	1.2764	0.6678
0.56	(110)(10)	5	6	F	5.5	0.6174	0.8356
0.48	(211)(30)	5	4	H	6.5	5.1595	0.0218
0.56	(211)(30)	5	4	H	6.5	3.6742	0.0993
0.47	(211)(21)	5	4	K	6.5	2.9116	0.1424
0.91	(110)(11)	5	6	H	6.5	0.9316	0.6213
0.73	(111)(20)	3	4	I	7.5	7.3017	0.0392
0.66	(221)(30)	5	2	K	7.5	6.6540	0.0132
0.50	(221)(31)	5	2	L	7.5	5.7567	0.0283
0.43	(211)(30)	5	4	K	7.5	5.2159	0.0371
0.62	(211)(21)	5	4	L	7.5	4.6911	0.0209
0.62	(211)(30)	5	4	I	7.5	3.6588	0.0100
0.60	(211)(30)	5	4	M	7.5	3.4434	0.0630
0.44	(211)(21)	5	4	K	7.5	2.3731	0.7151
0.87	(110)(11)	5	6	H	7.5	0.0000	2.2047
0.80	(211)(30)	5	4	K	8.5	5.5265	0.0473
0.70	(211)(21)	5	4	L	8.5	4.0603	0.0182
0.75	(211)(30)	5	4	M	8.5	3.8640	0.0573
0.64	(211)(21)	5	4	K	8.5	2.5067	0.9515
0.82	(211)(30)	5	4	M	9.5	4.0280	0.0394
0.75	(211)(21)	5	4	L	9.5	2.7342	1.1502
0.87	(211)(30)	5	4	M	10.5	2.7458	1.3075

Photoemission-Intensity in intermediate coupling
Transition from 5f11 to 5f10

Initial state eigenfunction: J = 7.5

0.9448	(111)(20)	3	4	I
-0.3221	(210)(21)	3	2	K
-0.0605	(210)(21)	3	2	L

Final state intensities:

					J	E (eV)	Intensity
0.49	(211)(21)	4	3	G	4.0	4.8811	0.0104
0.75	(111)(20)	4	5	G	4.0	4.7551	0.0208
0.54	(111)(20)	4	5	D	4.0	3.3008	0.0106
0.44	(111)(20)	4	5	G	4.0	2.6372	0.0545
0.46	(111)(20)	4	5	I	4.0	2.2272	0.0353
0.67	(111)(10)	4	5	F	4.0	2.0455	0.4282
0.60	(211)(30)	4	3	G	5.0	7.1482	0.0103
0.51	(211)(21)	4	3	H	5.0	5.3084	0.0103
0.69	(111)(20)	4	5	G	5.0	2.9146	0.5170
0.66	(111)(20)	4	5	I	5.0	2.3459	0.1515
0.55	(111)(10)	4	5	F	5.0	1.2227	0.5971
0.76	(110)(11)	2	3	H	6.0	9.6224	0.0206
0.69	(211)(20)	4	3	I	6.0	6.1007	0.0431
0.49	(211)(30)	4	3	I	6.0	4.4587	0.0682
0.48	(211)(30)	4	3	H	6.0	3.6650	0.1932
0.78	(111)(20)	4	5	G	6.0	2.4928	0.7614
0.62	(111)(20)	4	5	I	6.0	1.6130	0.8389
0.59	(211)(20)	4	3	I	7.0	7.1936	0.0126
0.61	(211)(30)	4	3	K	7.0	5.7211	0.0227
0.60	(211)(21)	4	3	L	7.0	5.0337	0.0605
0.58	(211)(30)	4	3	K	7.0	3.3772	0.2295
0.94	(111)(20)	4	5	I	7.0	1.3689	0.6717
0.63	(211)(21)	4	3	K	8.0	5.9756	0.0277
0.66	(220)(22)	4	1	L	8.0	4.4250	0.0726
0.53	(211)(30)	4	3	K	8.0	2.4575	0.9611
0.86	(111)(20)	4	5	I	8.0	0.0000	2.5550
0.91	(211)(21)	4	3	L	9.0	3.2565	1.2111
0.96	(211)(30)	4	3	M	10.0	3.8237	1.3341

Photoemission-Intensity in intermediate coupling
Transition from 5f12 to 5f11

Initial state eigenfunction: J = 6.0

0.9840	(110)(11)	2	3	H
0.1781	(200)(20)	2	1	I

Final state intensities:

					J	E (eV)	Intensity
0.56	(111)(20)	3	4	D	2.5	3.7434	0.0965
0.60	(111)(10)	3	4	F	2.5	2.4698	0.3523
0.73	(100)(10)	1	2	F	3.5	10.3786	0.0204
0.64	(210)(21)	3	2	G	3.5	6.6302	0.0109
0.48	(210)(20)	3	2	G	3.5	6.4282	0.0870
0.88	(111)(20)	3	4	D	3.5	4.2374	0.5379
0.57	(111)(20)	3	4	G	3.5	3.0553	0.4332
0.91	(111)(10)	3	4	F	3.5	2.7676	0.1819
0.85	(210)(11)	3	2	H	4.5	5.3503	0.0208
0.82	(111)(20)	3	4	G	4.5	3.2950	0.6332
0.57	(111)(20)	3	4	I	4.5	2.5091	0.1144
0.63	(111)(10)	3	4	F	4.5	1.0110	0.5702
0.73	(210)(20)	3	2	I	5.5	4.5136	0.1040
0.60	(210)(21)	3	2	H	5.5	3.9829	0.1203
0.69	(111)(20)	3	4	G	5.5	2.7110	0.7873
0.71	(210)(21)	3	2	H	5.5	1.2857	1.3333
0.74	(210)(20)	3	2	I	6.5	5.7052	0.0240
0.73	(210)(21)	3	2	K	6.5	4.1946	0.2096
0.98	(111)(20)	3	4	I	6.5	1.6326	0.8898
0.87	(210)(21)	3	2	K	7.5	3.3648	1.2584
0.94	(111)(20)	3	4	I	7.5	0.0000	2.7611
1.00	(210)(21)	3	2	L	8.5	4.6886	1.4192

Photoemission-Intensity in intermediate coupling
Transition from 5f13 to 5f12

Initial state eigenfunction: J = 3.5

1.0000	(100)(10)	1	2	F

Final state intensities:

					J	E (eV)	Intensity
0.89	(0)(0)	0	1	S	0.0	8.7145	0.0345
0.89	(110)(11)	2	3	P	0.0	3.5522	0.2155
1.00	(110)(11)	2	3	P	1.0	4.3351	0.3750
0.71	(200)(20)	2	1	D	2.0	5.5385	0.0743
0.75	(110)(11)	2	3	P	2.0	3.5826	0.6132
0.70	(200)(20)	2	1	D	2.0	1.5868	1.1875
1.00	(110)(10)	2	3	F	3.0	2.4778	0.8750
0.77	(110)(11)	2	3	H	4.0	4.3234	0.0307
0.60	(110)(11)	2	3	H	4.0	2.4245	1.0982
0.79	(110)(10)	2	3	F	4.0	0.4205	2.2461
1.00	(110)(11)	2	3	H	5.0	1.9572	1.3750
0.98	(200)(20)	2	1	I	6.0	4.2445	1.6946
0.98	(110)(11)	2	3	H	6.0	0.0000	3.1804

Table 3

transition	Initial state eigenfunction f^2, J = 4			Final state intensities f^1	
$f^2 \rightarrow f^1$	^3F	^3H	^1G	$^2F_{5/2}$	$^2F_{7/2}$
LS-limit	0	1	0	1.7143	0.2857
Pr^{3+}	-0.0300	0.9859	0.1645	1.8645	0.1355
Pa^{3+}	-0.0877	0.9489	0.3031	1.9547	0.0452
jj-limit	-0.1650	0.8650	0.4738	2.0000	0.0000

Table 4

Transition	$f^3 \rightarrow f^2$	$f^4 \rightarrow f^3$	$f^5 \rightarrow f^4$	$f^6 \rightarrow f^5$	$f^7 \rightarrow f^6$	$f^8 \rightarrow f^7$	$f^9 \rightarrow f^8$	$f^{10} \rightarrow f^9$	$f^{11} \rightarrow f^{10}$	$f^{12} \rightarrow f^{11}$	$f^{13} \rightarrow f^{12}$
HWHM γ (eV)	0.37	0.2	0.16	0.2	0.18	0.17	0.17	0.12	0.12	0.12	0.2
Asymmetry α	0.12	0.12	0.11	0.2	0.19	0.16	0.13	0.19	0.16	0.21	0.2
Scaling factor	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1

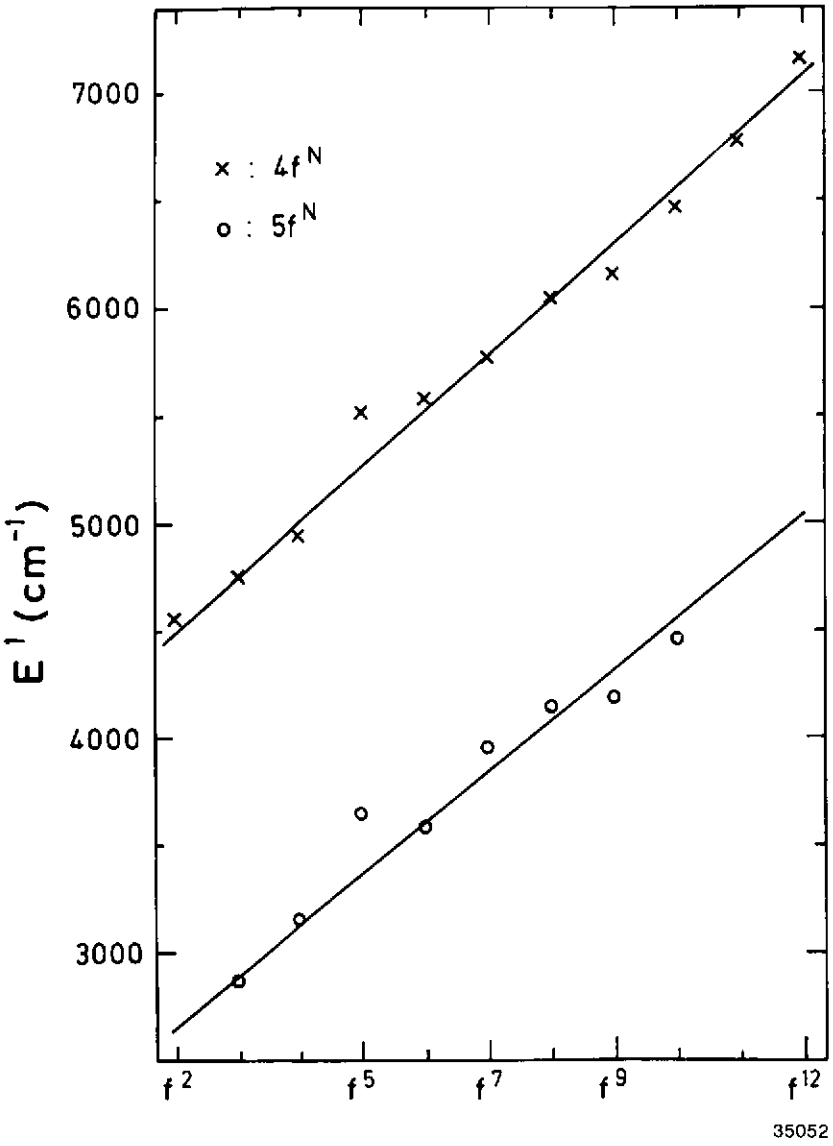
7000
6000
5000
4000
3000
E¹ (cm⁻¹)
× : 4f^N
o : 5f^N
f²
f⁵
f⁷
f⁹
f¹²
35052

Fig. 1

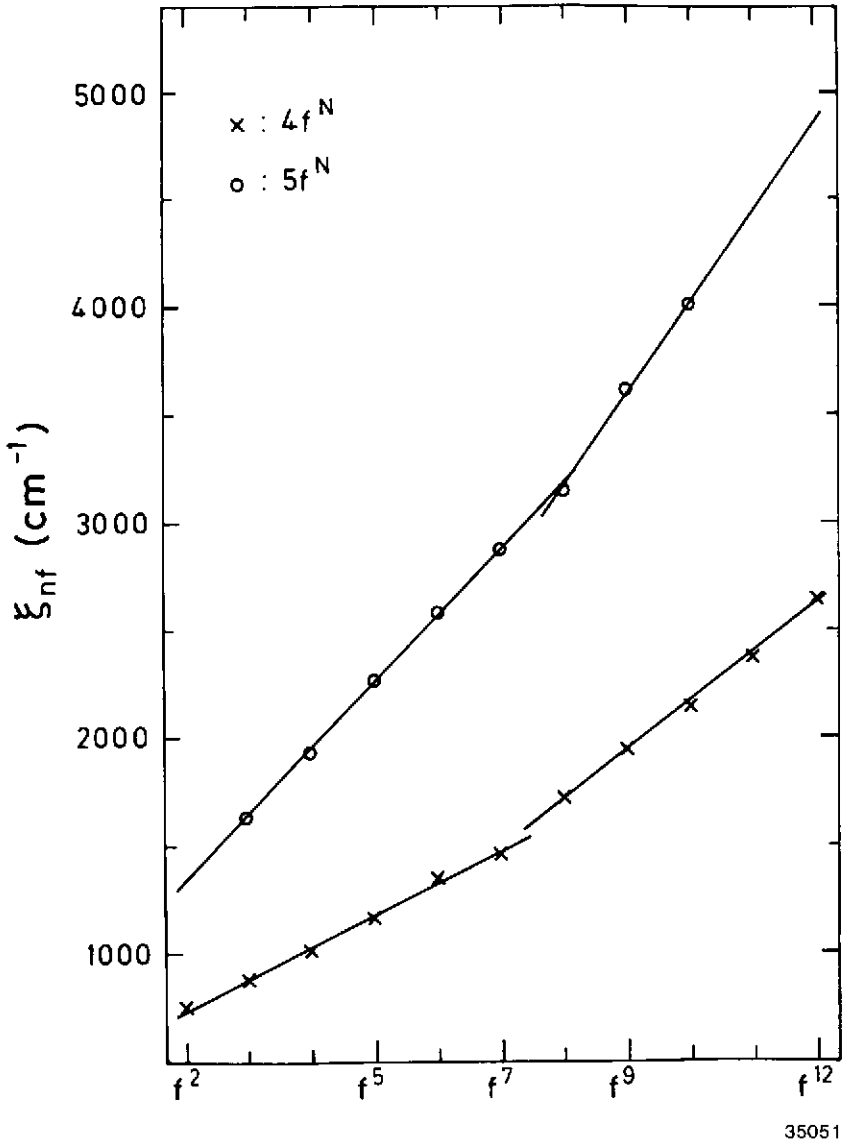
5000
4000
3000
2000
1000
ξnf (cm⁻¹)
× : 4f^N
o : 5f^N
f²
f⁵
f⁷
f⁹
f¹²
35051

Fig. 2

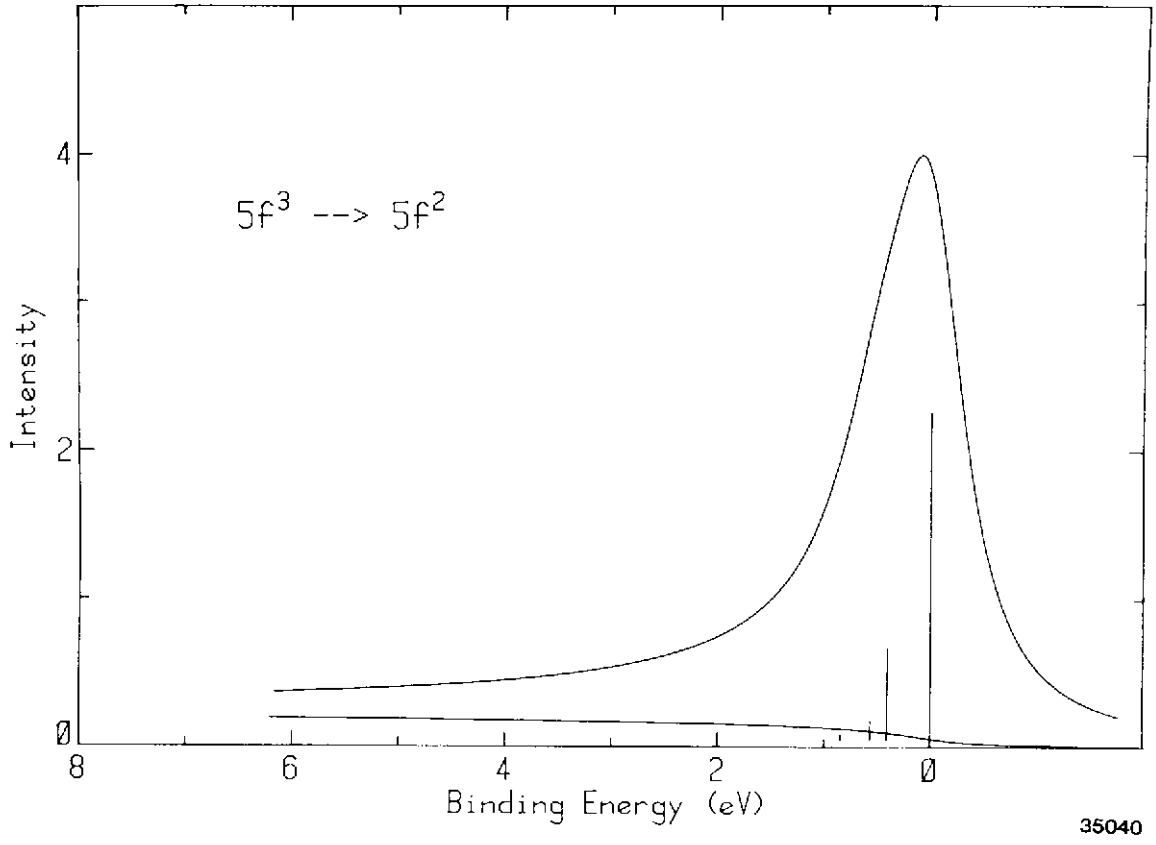
$5f^3$ --> $5f^2$
Intensity
4
2
0
8
6
4
2
0
Binding Energy (eV)
35040

Fig. 3a

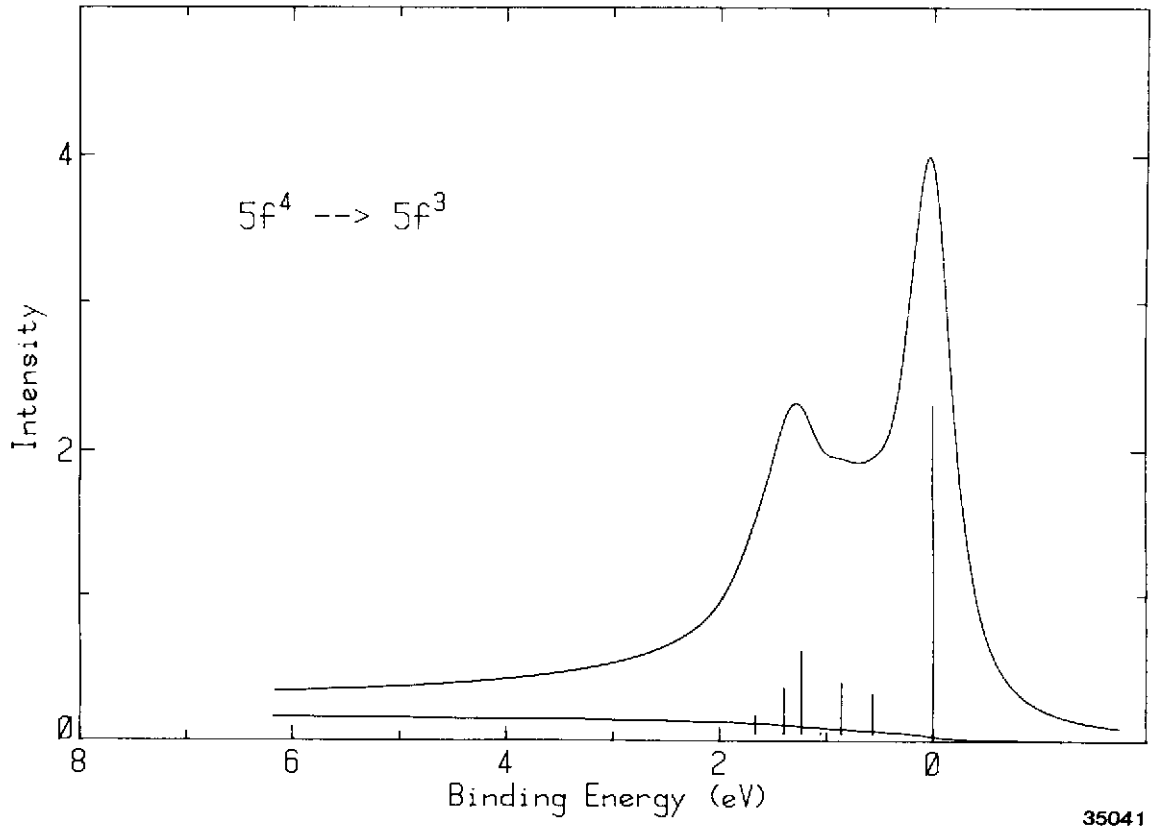
$5f^4$ --> $5f^3$
Intensity
4
2
0
8
6
4
2
0
Binding Energy (eV)
35041

Fig. 3b

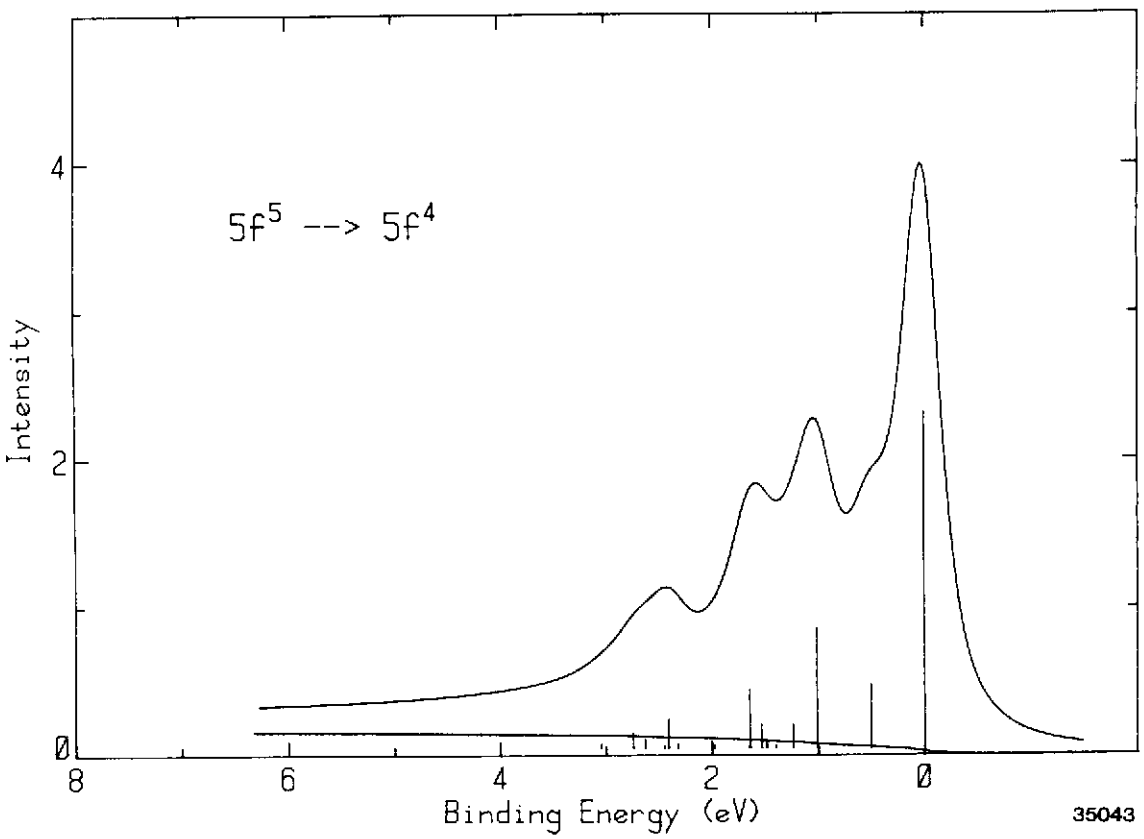
$5f^5$ --> $5f^4$
Intensity
4
2
0
8
6
4
2
0
Binding Energy (eV)
35043

Fig. 3c

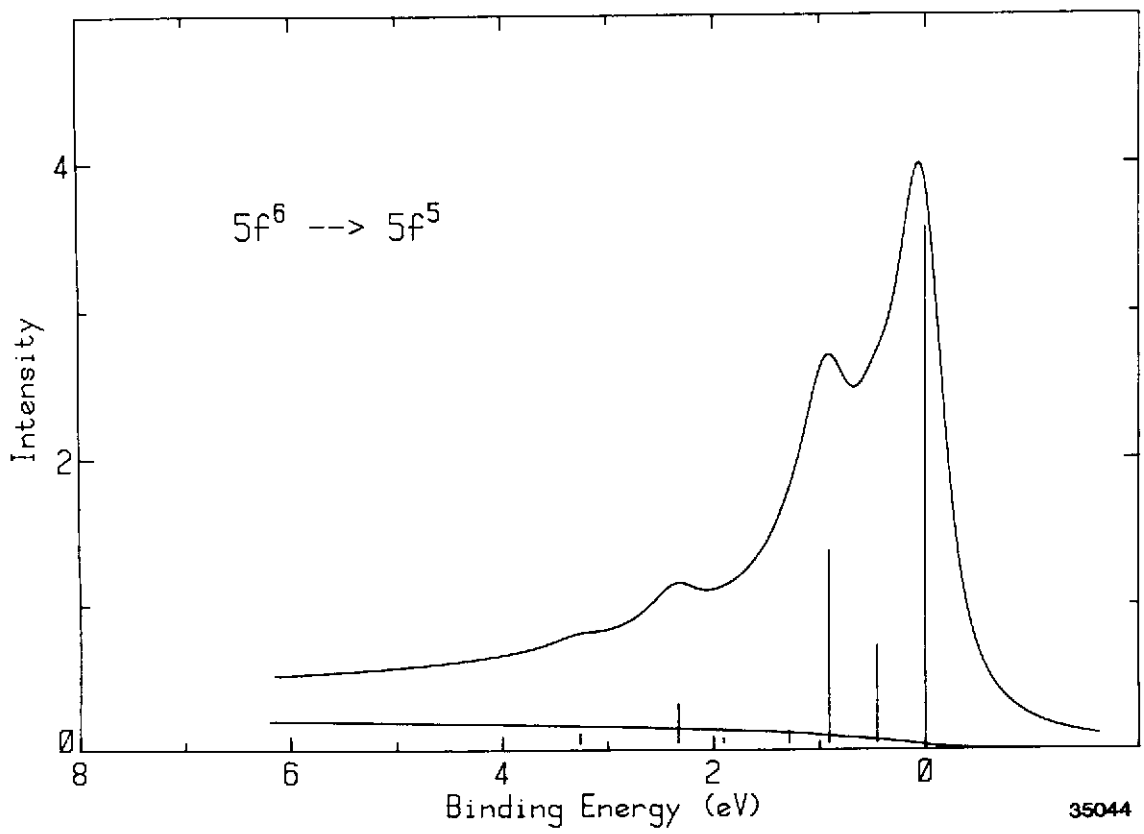
$5f^6$ --> $5f^5$
Intensity
4
2
0
8
6
4
2
0
Binding Energy (eV)
35044

Fig. 3d

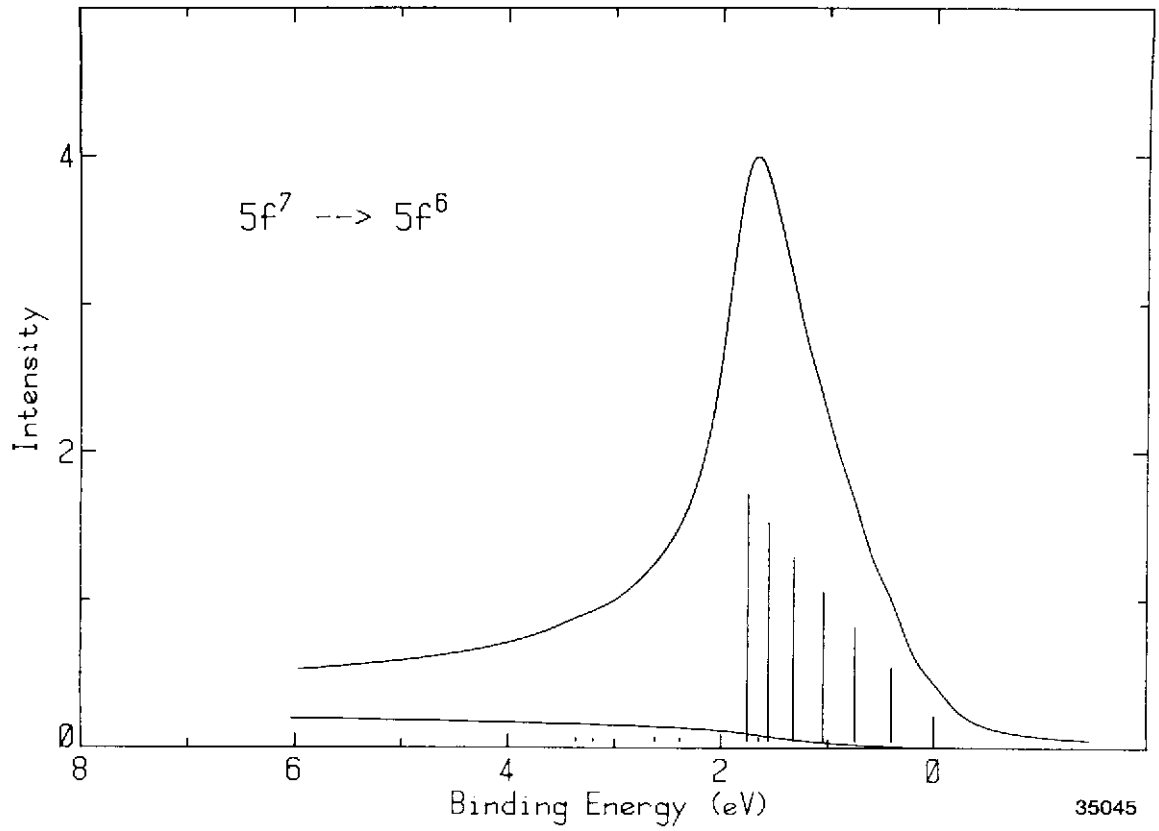
$5f^7$ --> $5f^6$
Intensity
4
2
0
8
6
4
2
0
Binding Energy (eV)
35045

Fig. 3e

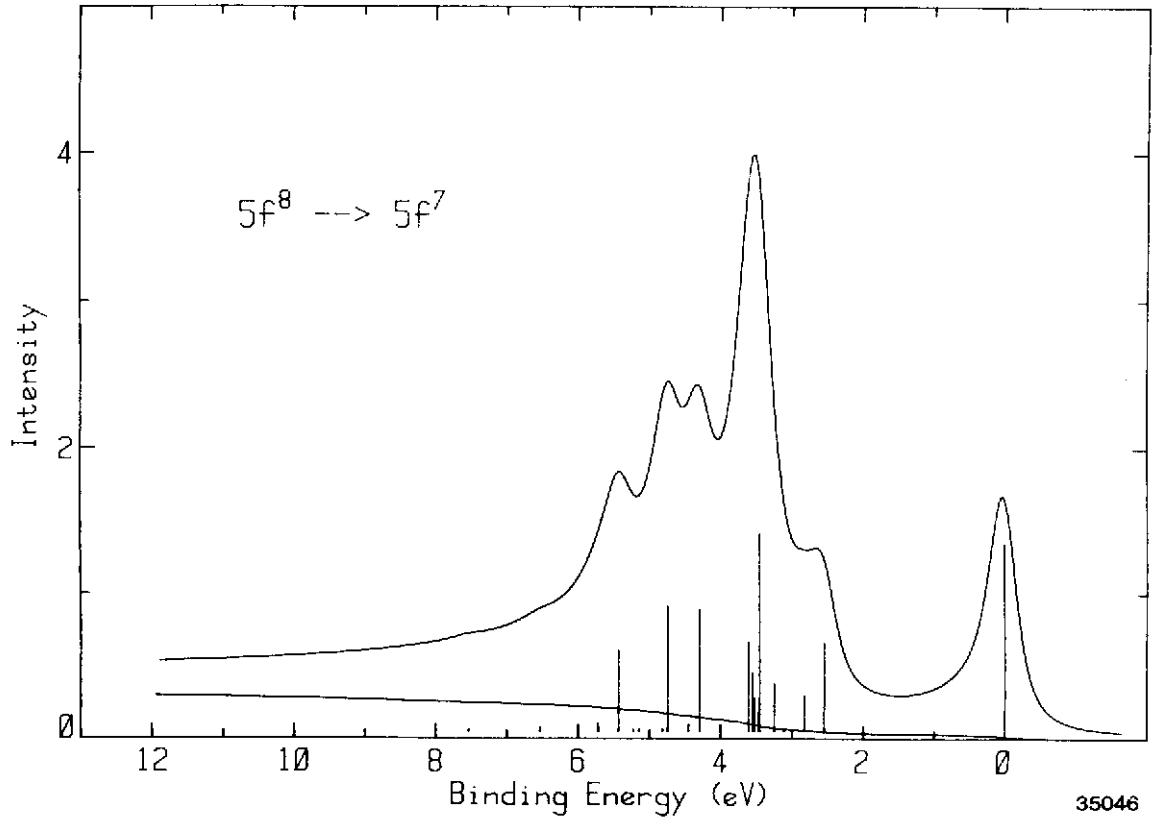
$5f^8$ --> $5f^7$
Intensity
4
2
0
12
10
8
6
4
2
0
Binding Energy (eV)
35046

Fig. 3f

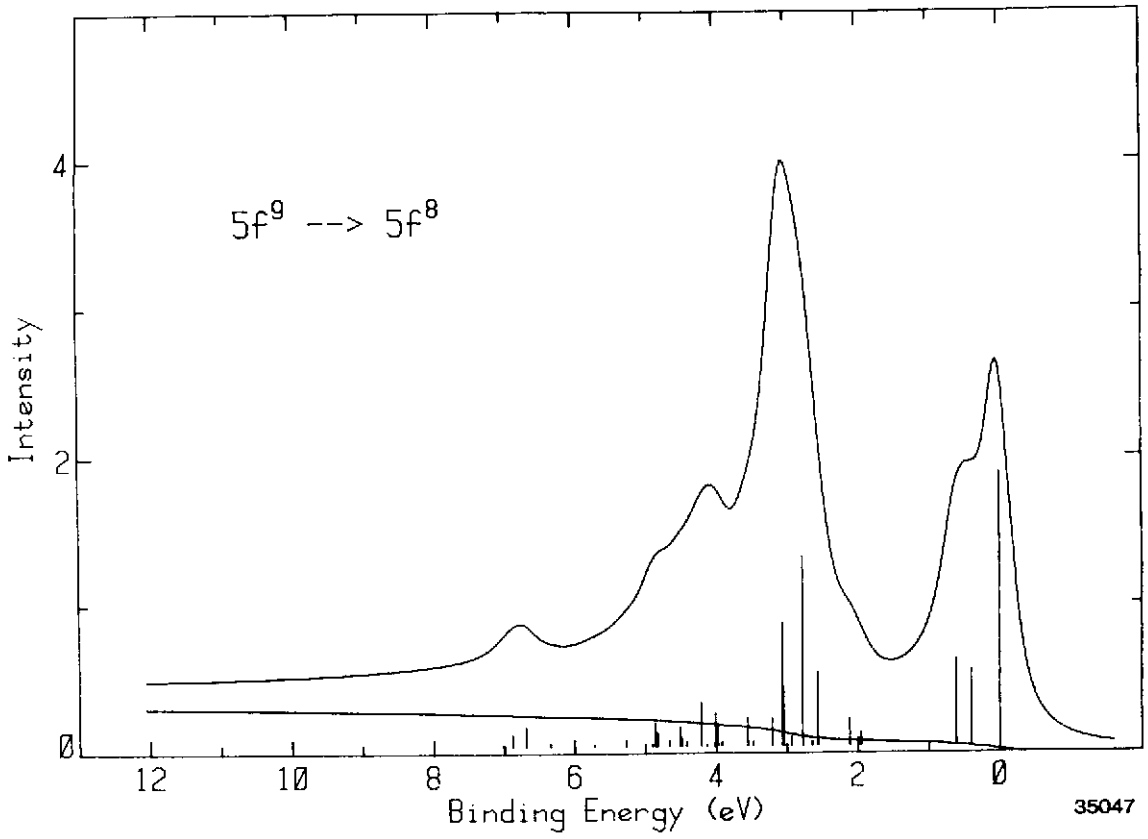
$5f^9$ --> $5f^8$
Intensity
4
2
0
12
10
8
6
4
2
0
Binding Energy (eV)
35047

Fig. 3g

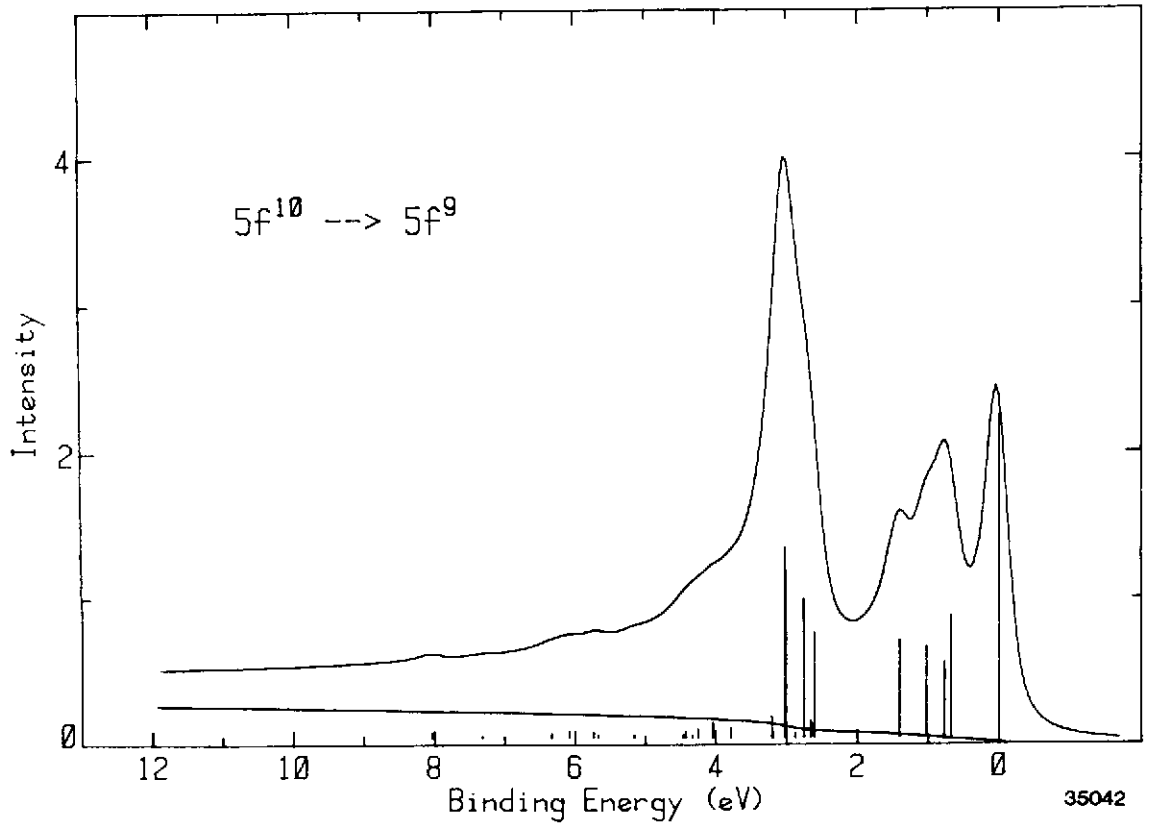
$5f^{10}$ --> $5f^9$
Intensity
4
2
0
12
10
8
6
4
2
0
Binding Energy (eV)
35042

Fig. 3h

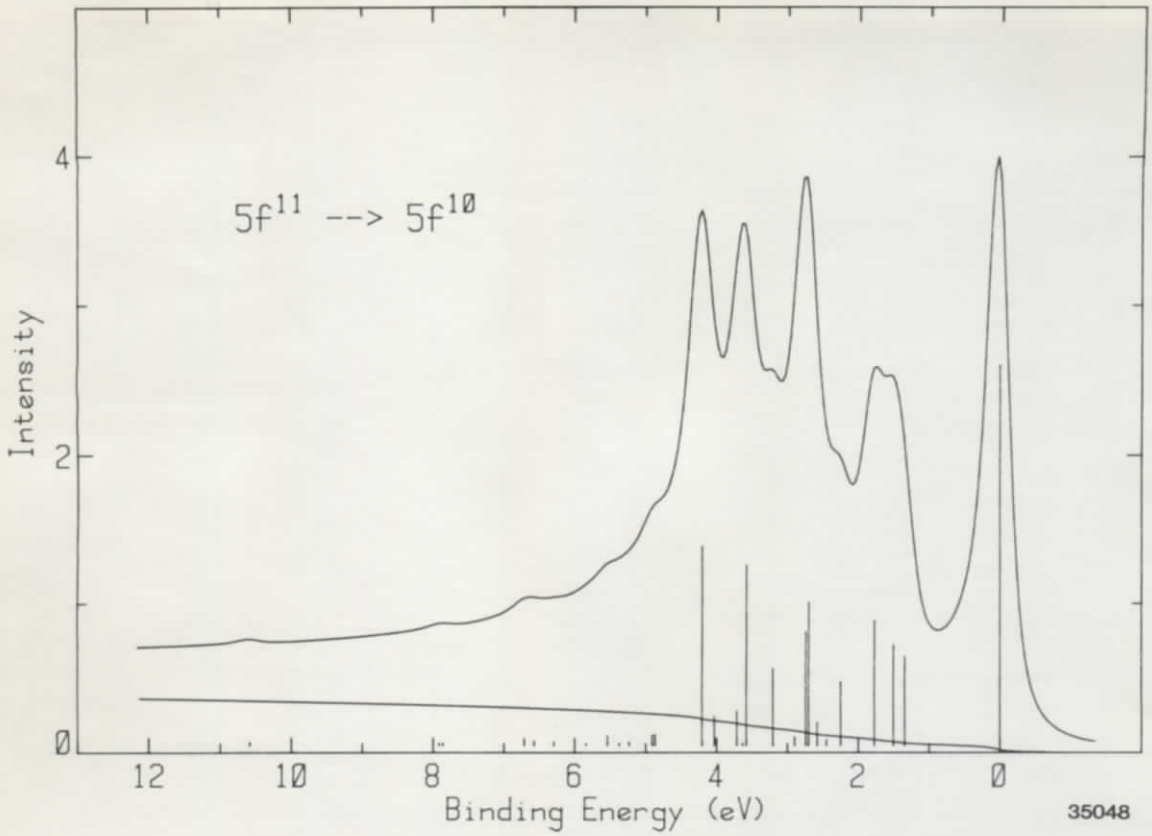
$5f^{11}$ --> $5f^{10}$
Intensity
4
2
0
12
10
8
6
4
2
0
Binding Energy (eV)
35048

Fig. 3i

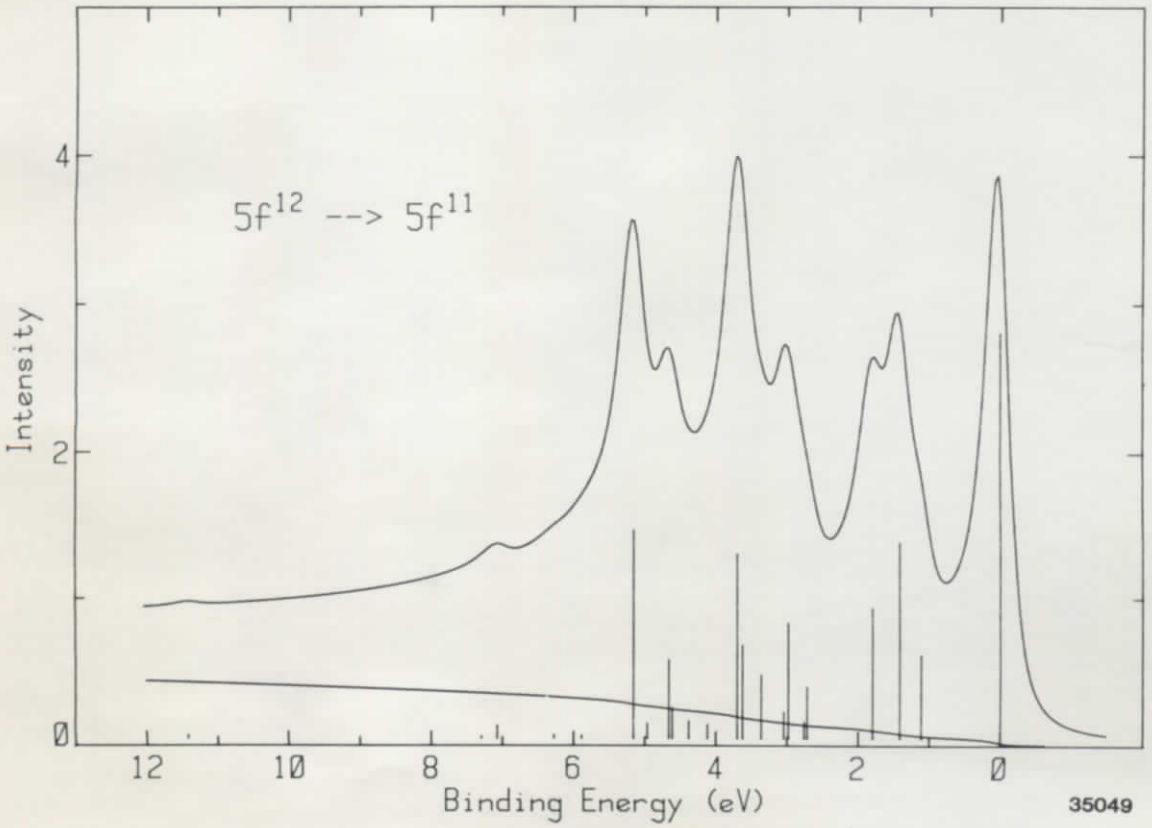
$5f^{12}$ --> $5f^{11}$
Intensity
4
2
0
12
10
8
6
4
2
0
Binding Energy (eV)
35049

Fig. 3j

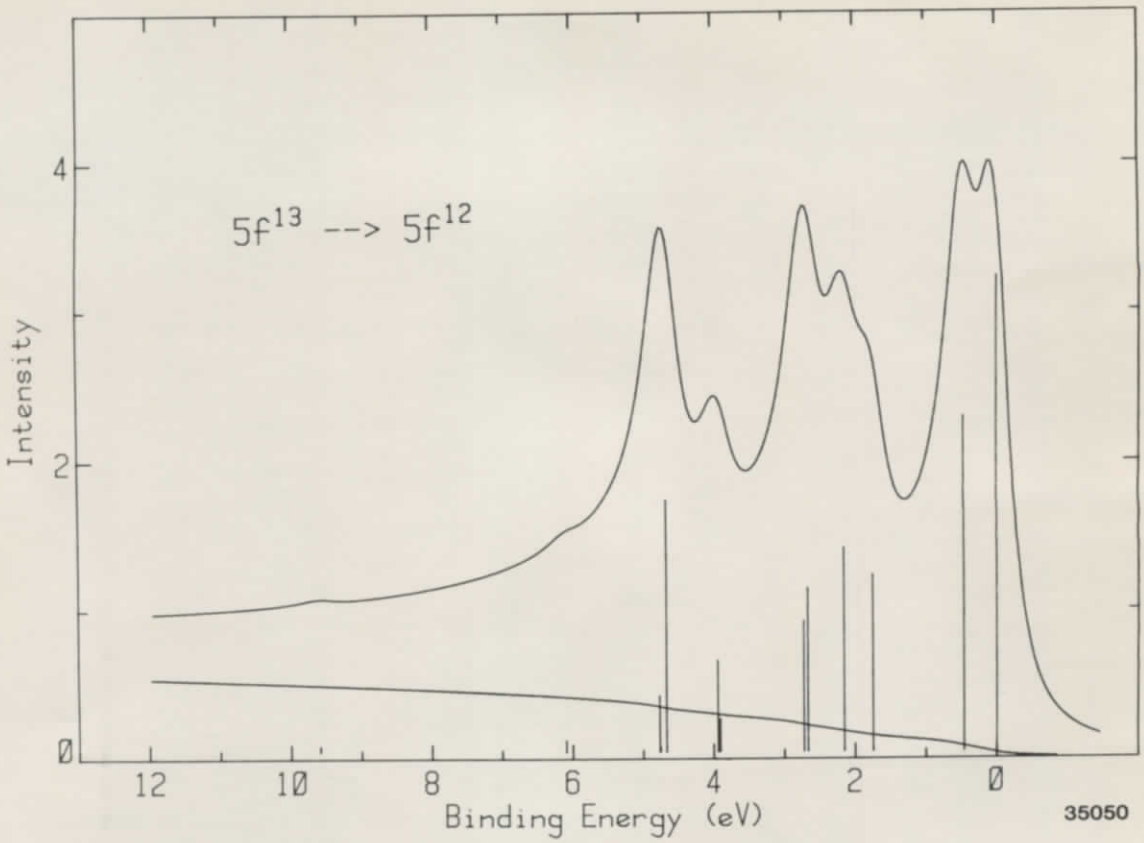

Fig. 3k