#### INVESTIGATION OF DELAYED NEUTRON EMISSION THROUGH NEUTRON AND GAMMA-RAY SPECTROSCOPY

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### Abstract

Fast radiochemical separations have permitted detailed and high resolution measurements of neutron and  $\gamma$ -ray spectra from several delayed neutron emitting systems. The apparent discrete line structure in delayed neutron spectra, high intensity neutron branching to excited states in decay of intermediate levels in the emitter, and the peaking in the  $\beta$ -decay intensity to regions well above the neutron binding energy, indicate persistence of distinct nuclear structure effects at excitation energies of 5-7 MeV in the emitter nuclides.

### Introduction

The phenomenon of  $\beta^-\text{-del}$  ayed neutron emission provides an interesting tool for studying nuclear properties at intermediate excitation energies in neutron rich nuclei far off the stability line. The fact that most of the high resolution delayed neutron spectra so far known^1-3) show prominent line structure arouses considerable interest in determining nuclear level densities at excitation energies above the neutron binding energy (B\_n), as well as investigation of  $\beta^-\text{-strength}$  functions.

### Experimental delayed neutron spectra

We have studied extensively the delayed neutron emission from four precursor nuclides, i.e.,  $^{85}$ As,  $^{87}$ Br,  $^{135}$ Sb and  $^{137}$ I, two each forming characteristic pairs in the lightand heavy mass regions in fission. The members of each pair differ only by a pair of protons. The corresponding neutron emitters have N = 51 or 83, respectively, leading to nuclei with major closed shells after neutron emission.

The nuclides  $^{87}\text{Br}$  and  $^{137}\text{I}$  have a relatively small energy window ( $Q_{\text{B}}\text{-B}_{\text{n}}$ ) for neutron emission, whereas  $^{85}\text{As}$  and  $^{135}\text{Sb}$  have large energy windows, allowing neutron transitions not only to the ground state but also to several excited states in the final nuclei. In the latter cases the characteristics of delayed neutron emission should be revealed most clearly.

The neutron spectra were measured after rapid chemical separation<sup>4-6</sup>) with commercially available <sup>3</sup>He ionization chambers<sup>7</sup>) with a resolution of 12 keV and 19 keV for thermal and 1 MeV neutrons, respectively. The improved resolution has resulted from minimization of effects due to pulse summing, detection of scattered neutrons, and microphonics<sup>8</sup>), and is most evident in the low-energy parts of the neutron spectra.

The pulse-height distribution from <sup>87</sup>Br decay, obtained directly by summing data from 280 experiments is shown in Figure 1 (upper part); the neutron spectrum obtained after correction for the detector

response function and for thermal neutrons is shown in the lower part. The neutron spectra of <sup>85</sup>As, <sup>135</sup>Sb and <sup>137</sup>I acquired with the same procedure are given in Figures 2-4. For reasonable counting statistics, spectra from 90 (As), 210 (Sb) and 375 (I) experiments, respectively, were accumulated. (Note that the published spectra<sup>2</sup> of <sup>85</sup>As and <sup>135</sup>Sb have not been corrected for detector response).

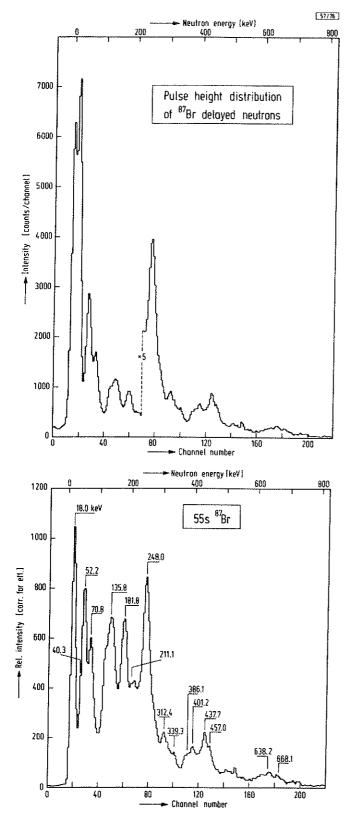
All four spectra show prominent neutron line structure containing the principle part of the neutron intensity (>60 %). While in the case of  $^{87}\mathrm{Br}$  and  $^{137}\mathrm{I}$ , the maximum neutron energy observed is in agreement with the energy range ( $Q_8^-B_n$ ) available for neutron emission, a dominant feature of the delayed neutron spectra of  $^{85}\mathrm{As}$  and  $^{135}\mathrm{Sb}$  is the absence of appreciable neutron intensity above 1.6 MeV (As) and 2.0 MeV (Sb) though ranges of about 4.7 MeV and 4.1 MeV, respectively, are possible for the neutron energies.

#### Neutron emission to excited states

Population of excited states by neutron emission was first observed through the complexity of decay curves of  $\gamma$ -ray peaks in the arsenic fraction of fission products  $^9,10$ ). The neutron branching into excited states of  $^{84}$ Se was confirmed by recent experiments at the LOHENGRIN fission fragment spectrometer where mass separated samples at A = 85 showed  $\gamma$ -rays emitted from excited states in  $^{84}$ Se  $^{11}$ ). In a similar way, neutron decay into excited states of  $^{134}$ Te has been observed in the mass chain 135. The same  $\gamma$ -rays were seen following  $\beta$ -decay of the respective parents of the neutron decay final nuclei in the neighbouring mass chain, i.e.  $^{84}$ As( $\beta^*$ ) $^{84}$ Se and  $^{134}$ Sb( $\beta^*$ ) $^{134}$ Te. From the intensity ratios of such  $\gamma$ -rays in the two mass chains,partial neutron emission probabilities,  $P_{1}^{1}(\gamma)$ , to certain levels i in the final nucleus were determined. These data indicate strong neutron feeding of excited states in both neutron emitting systems: 59  $\pm$  11 % for  $^{85}$ Sh and 53  $\pm$  12 % for  $^{135}$ Sb $^{11}$ ). From those  $P_{1}^{1}$  values one deduces peaking in the  $\beta^-$ -strength near 6-7 MeV in  $^{85}$ Se and  $^{135}$ Te, indicating high selectivity in  $\beta^-$ -decay to this energy region.

### Spectral analysis

The line structure in the neutron spectra of  $^{87} \mathrm{Br}$  (Figure 1) and of  $^{135} \mathrm{Sb}$  (Figure 4) has been analyzed independently by two peak fitting routines, SAMPO¹²) and NEUTRN¹³). Both programs utilize gaussian or modified gaussian representations for a single line and fit regions of the spectrum with linear or parabolic representations of underlying continua. The results from both fitting procedures were in very good agreement with respect to peak centroids and areas of all but the weakest lines. It was found that in the case of  $^{87} \mathrm{Br}$  about



### Figure 1

Neutron spectrum from <sup>87</sup>Br decay. Experimental pulse height distribution (upper part), neutron spectrum (lower part).

85% and in the case of  $^{135}$ Sb about 70 % of the neutron intensity was accounted for by the fitted lines.

# Level density and $(n-\gamma)$ competition in $^{67}$ Kr

In the case of  $^{87}Br$ , all neutrons are emitted to the ground state of  $^{86}Kr$  and an analysis of the neutron spectrum leads directly to an estimate of the density of levels strongly populated by  $\beta^-$ -decay. In the energy range o < E $_{n}$  < 300 keV, the neutron intensity can be accounted for by 14 transitions, implying a density of about 47 (MeV) $^{-1}$  at 5.7 MeV in  $^{87}Kr$ . This can be compared to the values of 120 (MeV) $^{-1}$  and 200 (MeV) $^{-1}$  for (1/2 $^-$ , 3/2 $^-$ ) levels and (1/2 $^-$ -5/2 $^-$ ) levels, respectively, obtained with the Gilbert and Cameron formulae  $^{14}$ ).

Through study of the  $\gamma$ -ray spectrum from <sup>87</sup>Br decay<sup>15,16</sup>) 11 levels have been found with E\*>Bn. For 8 of these, correspondences with level energies from the neutron lines have been established using the recently measured value of 5515.2  $\pm$  1.0 keV<sup>17</sup>) for the neutron binding energy (Table 1). For these levels the value of 2.8 was obtained for  $(\Gamma_{\gamma}/\Gamma_{\eta})$  and life times of the levels are estimated to lie in the range (0.4 - 2)· $10^{-16}$  sec.

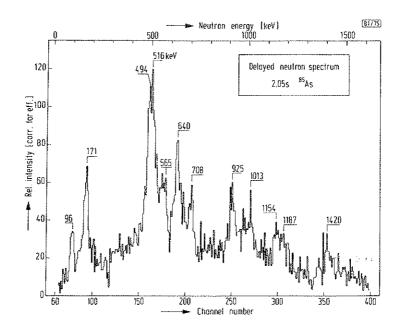
# Definition of intermediate states in <sup>135</sup>Te

In the case of <sup>135</sup>Sb, where a substantial fraction of the neutrons emitted from intermediate levels in <sup>135</sup>Te leads to excited states in the final nucleus, unfolding of the neutron spectrum is necessary in order to determine individual neutron emitting levels, as well as the  $\beta$ -strength function. Through comparison of the neutron peak energies (E<sub>n</sub>) with level energies in the final nucleus (E<sub> $\ell$ </sub>), we conclude that neutron decay from certain special levels in the emitter may feed different states in the final nucleus <sup>134</sup>Te. We find 20 correlations for <sup>135</sup>Sb, where  $(E_{n,1}-E_{n,2})=(E_{\ell,2}-E_{\ell,1})$ , giving the energies of the neutron emitting levels in <sup>135</sup>Te within less than ± 5 keV uncertainty. However, arguments concerning spin and parity of the levels involved, the log ft-values of the β-transitions preceeding neutron emission, and the restriction to the single placement of each neutron line, reduces the final number of correlations to 12. This results in 7 neutron emitting levels in  $^{135}{\rm Te}$  (Figure 5). The number of correlated neutron lines is significantly larger than that resulting from probability calculations including all energetically possible combinations.

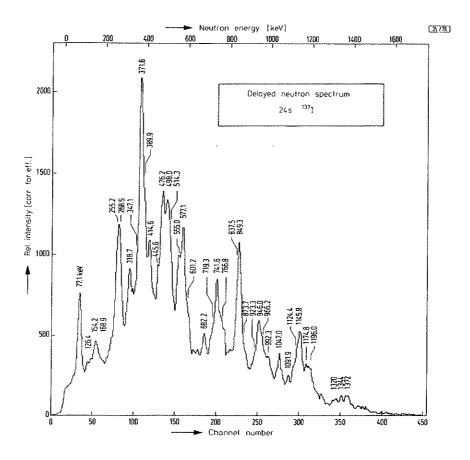
The partial neutron emission probabilities to each of the excited states in  $^{134}$ Te obtained by summing the intensities of the neutron transitions shown in Figure 5 are in agreement with the respective partial neutron emission probabilities from  $\gamma$ -ray measurements $^{11}$ ). On the basis of this intensity balance, the non-correlated neutron lines must mainly populate the ground state in  $^{134}$ Te.

From the absolute intensities of the neutron transitions, and the known  $P_n\text{-value}$  for the  $^{135}\text{Sb}$  precursor  $^{18}$ ),  $\beta^-$  intensities to the neutron emitting levels were determined. The corresponding log ft-values were found to be in the range of 3.6 - 5.3 (Figure 5), consistent with allowed transitions which are expected to dominate the  $\beta^-$ -decay of  $^{135}\text{Sb}$ .

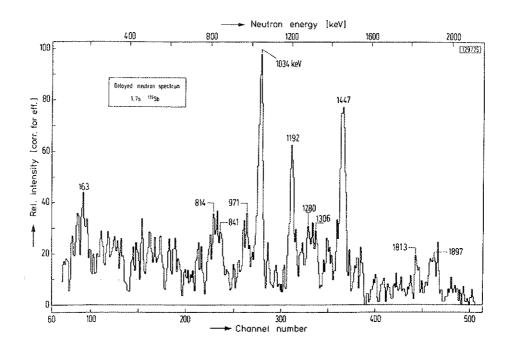
Assuming a ground state spin and parity of  $7/2^+$  for  $^{135}$ Sb and the spins and parities of the final states  $^{19}$ ) in  $^{134}$ Te as shown in Figure 5, transmission coefficient ratios derived from optical model



 $\frac{\text{Figure 2}}{\text{Neutron spectrum from }^{85}\text{As decay after correction for detector efficiency and thermal neutrons.}}$ 



 $\frac{\text{Figure 3}}{\text{Neutron spectrum from }^{137}\text{I decay after correction of detector efficiency and thermal neutrons.}}$ 



 $\frac{\text{Figure 4}}{\text{Neutron spectrum from $^{135}$Sb decay after correction for detector efficiency and thermal neutrons.}}$ 

calculations  $^{20}$ ) for neutron waves implying allowed  $\beta^-$ -transitions were compared to the respective experimental neutron intensities. For all seven neutron emitting levels in  $^{135}\text{Te}$  a search was made to define those spin assignments which would provide the best agreement between neutron branching ratios and optical model transmission coefficient ratios. Table 2 shows a comparison of experimental and calculated neutron intensities, the latter normalized in each case to a d-wave neutron transition. It is immediately evident that a serious discrepancy exists relative of high-energy neutron transitions (E\_n  $\geq$  1.5 MeV) regardless to wave type: such transitions were generally unobserved in the experimental

neutron spectrum. For the four transitions assigned to the decay of the 6.90 MeV level, good agreement between experimental and calculated intensities was found for the two lowest energy branches, but the experimental intensities for the higher energy transitions are seen to decrease rapidly with energy compared to the calculated values. For the remaining six neutron emitting levels only two transitions for each could be assigned. For all but one, the intensities of competing s-wave transitions appear low compared to those expected from the transmission coefficient ratios. This result is consistent with the small value for the total neutron intensity to the  $4^{+}$ level in  $^{134}$ Te relative to those to the neighbouring  $2^{+}$  and  $6^{+}$  levels.

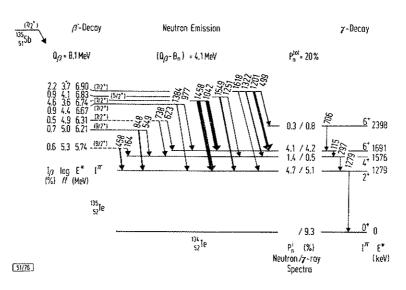


Figure 5
Decay scheme proposed for the  $^{135}Sb(\beta^-)^{135}Te(n,\gamma)^{134}Te$  system

 $\frac{ \mbox{Table 1}}{\mbox{Comparison of level energies in $^{87}$Kr derived from neutron and $\gamma$-ray spectra}$ 

E <sub>l,n</sub> (	E <sub>ℓ,n</sub> (ΔE <sub>ℓ,n</sub> )		ΔΕ <sub>ε,γ</sub> )a	ΔE/σ <sub>T</sub>  b		
5533.4	(1.8)	5530.8	(0.3)	1.4		
		5543.6	(0.8)			
5556.0	(1.8)					
5568.0	(2.5)					
5586.8	(2.0)	5588.6	(1.0)	0.8		
5596.0	(2.8)	5595.1	(0.5)	0.3		
5637.7	(3.0)	5635.1	(0.5)	0.8		
5652.6	(2.1)					
5664.4	(2.7)	5659.3	(8.0)	1.8		
		5673.0	(0.4)			
5686.4	(3.5)	5686.9	(0.4)	0.1		
5699.1	(3.3)					
5728.8	(2.6)	5720	(3)	2.2		
5766.1	(3.6)					
5774.4	(2.7)					
		5794.2	(0.8)			
5831.2	(2.6)	5821	(5)	1.8		
5858.4	(3.3)					
5906.0	(3.3)					
5921.1	(3.3)					
5927.3	(3.3)					
5958.0	(2.6)					
6160.8	(4.0)					
6191.1	(4.3)					

 $<sup>^{\</sup>rm a}$ Based on the analysis of F.M. Nuh et al. $^{15}$ ) and Tovedal and Fogelberg $^{16}$ ).

bDefined as 
$$|E_{\ell,n}-E_{\ell,\gamma}|/(\sigma_{\ell,n}^2+\sigma_{\gamma}^2)^{1/2}$$
.

The same comparison of experimental and calculated neutron branches was performed for  $^{85}\mathrm{As}$  and yielded similar results. The Ph-values from neutron and  $\gamma$ -ray data agreed within the experimental errors. Overall agreement was found between experimental and calculated low-energy neutron branches involving mainly p-wave neutrons, whereas the calculated high-energy neutron transitions (En-1.4 MeV) were not found in the experimental neutron spectrum.

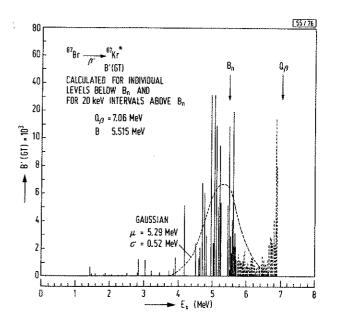
### Beta-strength functions of 87Br and 135Sb

A  $\beta^-$ -strength function for decay of  $^{87}{\rm Br}$  has been constructed by combining the decay scheme  $^{15}$ ) with the neutron measurements. For excitation energies in  $^{87}{\rm Kr}$  in the range  ${\rm B_n} < {\rm E^x} < ({\rm B_n} + 260)\,{\rm keV}$ , we have assumed that the neutron transitions found in our analysis account for the entire neutron intensity. At higher energies, the spectral analysis is not complete and we have determined  $\beta^-$ -intensities to 20-keV intervals in this region.

The quantity calculated was the reduced Gamow-Teller transition probability

$$B'(GT) = \frac{6260(g_V/g_A)^2}{ft_{1/2}}$$
;

and we have used the value of 7.06 MeV for  $Q_{\beta}^{-21}$ ). The resultant distribution is shown in Figure 6. The figure is dominated by a pronounced peak centered at 5.3 MeV which, by considering the cumulative strength above 3.0 MeV, is well represented by a gaussian distribution with  $\sigma$  = 0.52 MeV. The strength located near 2.7 - 3.0 MeV represents transitions to the lowest odd parity levels identified in the decay scheme<sup>15</sup>). The sharp rise in B'(GT) above 6.3 MeV (E\_{n} = 0.8 MeV) is dependent upon the uncertainty in  $Q_{\beta}$ , butan increase as  $Q_{\beta}$  is approached must occur. When smoothened, the variations in B'(GT) shown in Figure 6 are consistent with the strength function measured by Aleklett et al.  $^{22}$ ) using a total absorption spectrometer.



# Figure 6

Experimental \$\begin{align\*} -strength function for \$^{87}\$Br decay

The  $\beta^-$ -strength function for  $^{135}\text{Sb}$  decay to levels in the range  $B_n$  to  $Q_\beta$  in  $^{135}\text{Te}$  has been constructed using the levels shown in Table 2 with the assumption that the non-correlated neutron intensity must directly populate the ground state in  $^{134}\text{Te}$ . In Figure 7 this strength function is shown and we indicate by the broken line, the crude strength function in the energy range 0 - 6 MeV inferred by analysis of the  $\gamma$ -ray spectrum from  $^{135}\text{Sb}$  decay. The strong concentration of  $\beta^-$ -strength near 7 MeV is quite evident and the ratio of strength contained in levels near this energy is roughly a factor of  $10^2$ that contained in levels near 5.5 MeV.

# Conclusions

The structure demonstrated in the four neutron spectra shown in Figures 1 - 4 seems considerably beyond that which can be expected from simple statistical model considerations and points to appreciable selectivity in  $\beta^-\text{-decay}$  to levels in the range 5 - 7 MeV in the emitter nuclides²). In addition, the low intensity of high-energy neutrons

Table 2 Neutron transitions in decay of intermediate levels in  $^{135}$ Te: Comparison of experimental neutron intensities with those calculated from optical model transmission coefficients $^{20}$ )

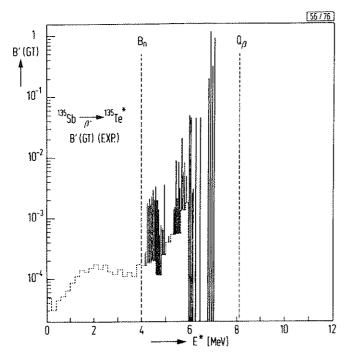
Veutron er E* MeV a	nitting leve (E <sup>*</sup> -B <sub>n</sub> )  keV	el in <sup>135</sup> T I <sup>n</sup> i	e <sup>£</sup> n- wave	Level in <sup>134</sup> Te I <sup>π</sup> f	energ	transition y E <sub>n</sub>  keV  <sup>b</sup> d <sup>C</sup> observed		ensity (relative calculated
5.74	1739 <u>+</u> 4	(9/2+)	<b>d</b> 3/2	6 <sub>1</sub> <sup>+</sup>	47	-	< 5	0
			s	4+	162	164±3	11	58
			$d_{5/2}$	z <sup>+</sup>	459	458 <u>+</u> 2	16	16 <sup>e</sup>
			$g_{9/2}$	0+	1739	-	<3	6
6.13 2126±4	2126+4	(9/2 <sup>+</sup> )	d <sub>3/2</sub>	6 <sup>‡</sup>	435	-	<5	3
			s `	4+	550	549+3	13	29
			d <sub>5/2</sub>	2+	847	848 <u>+</u> 3	20	20 <sup>e</sup>
		99/2	0+	2126	-	<7	5	
6.31 2314±5	(7/2 <sup>+</sup> )	d <sub>5/2</sub>	62	85	-	<5	0	
		d <sub>5/2</sub>	61	623	623+3	11	11 <sup>e</sup>	
		s -/	4+	738	 738 <u>+</u> 3	11	30	
		d <sub>3/2</sub>	2+	1035	_	<5	12	
		g <sub>7/2</sub>	0+	2314	-	<5	3	
6.67 2666 <u>+</u> 5	(7/2 <sup>+</sup> )	d <sub>5/2</sub>	62	268	_	<5	2	
		d <sub>5/2</sub>	6 <del>1</del>	975	977+2	25	25e	
		s -, -	4+	1090	-	<5	34	
		d <sub>3/2</sub>	2+	1387	1384+4	14	20	
		97/2	0+	2666	-	<1.5	6	
6.74 2735 <u>+</u> 3	(7/2 <sup>†</sup> )	d <sub>5/2</sub>	62	337		<5	9	
		d <sub>5/2</sub>	61	1044	1042+2	87	87 <sup>e</sup>	
			s	4+	1159		<5	105
		d <sub>3/2</sub>	2+	1456	1458+3	100	72	
		g <sub>7/2</sub>	0+	2735	_	<0.5	19	
6.83 2827 <u>+</u> 5	(5/2 <sup>+</sup> )	97/2	62	430	<b>*</b>	<5	0	
		g <sub>7/2</sub>	61	1136	_	<5	2	
		$d_{3/2}$	6 <sup>+</sup> 4 <sup>+</sup>	1251	1251+4	10	10 <sup>e</sup>	
			s	2 <sup>+</sup>	1548	 1549+3	27	20
		$d_{5/2}$	0+	2827	-	<0.2	23	
6.90 289 <u>6+</u> 4	2896 <u>+</u> 4	(7/2 <sup>+</sup> )	d <sub>5/2</sub>	62	498	499+3	12	12
			d <sub>5/2</sub>	61	1202	1201+2	51	51 <sup>e</sup>
			S	6 <sup>+</sup> <sub>2</sub> 6 <sup>+</sup> <sub>1</sub> 4 <sup>+</sup>	1320	1322+4	24	56
		$d_{3/2}$	2 <sup>+</sup>	1617	1618+5	10	42	
			97/2	0+	2896	**	<0.6	13

 $<sup>{}^</sup>aE^* = (E_n + B_n)$  with  $B_n = 4.0$  MeV.  ${}^bKinetic$  energy corrected for recoil energy.

 $<sup>^{\</sup>rm C}$ According to level scheme Figure 5.

 $<sup>^{</sup>m d}$ From optial model transmission coefficients $^{20}$ ).

eNormalized to observed neutron transition.



### Figure 7

Experimental B-strength function for 135Sb decay

cannot be attributed solely to angular momentum factors and demonstrates selectivity in neutron emission as well. We conclude that some structure effects must be accounted for in a successful description of delayed neutron emission.

It has already been suggested that this structure in delayed neutron spectra may be associated with decay to the antianalogue states  $^2$ ). The same conclusion is obtained from the M1  $_{\rm Y}$ -decay of isobaric analogue states to the antianalogue, core polarization and spin-flip states in this energy range  $^{2.3}$   $^{2.4}$ ) from another point of view, Shihab-Eldin et al.  $^{2.5}$ ) have shown that simple shell-model calculations locate the configurations in emitter nuclides reached by allowed  $_{\rm S}^-$ -decay in the same energy range.

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#### References

- S. Shalev and G. Rudstam, Nucl. Phys. <u>A</u> 230, 153 (1974).
- 2) H. Franz et al., Phys. Rev. Lett. 33, 859 (1974).
- G. Rudstam and S. Shalev, Nucl. Phys. <u>A235</u>, 397 (1974).
- 4) H. Folger et al., Radiochem. Radioanal. Lett.  $\underline{1}$ , 185 (1969).
- W. Eckhardt et al., Z. Anal. Chem. 226, 71 (1967) and H. Ohm, Diploma Thesis, Universität Mainz (1975).
- 6) N. Trautmann, contribution to this conference.
- Seforad-Applied Radiation Ltd., Emak Hayarden, Israel.
- 8) H. Franz et al., to be published.
- 9) H. Franz et al., Angew. Chem. 83, 902 (1971).
- 10) J.-V. Kratz et al., Nucl. Phys. A250, 13 (1975).
- 11) K.-L. Kratz et al., submitted to Phys. Rev. Lett.
- 12) J.T. Routti and S.G. Prussin, Nucl. Instr. Methods <u>72</u>, 125 (1969).
- 13) H. Ohm et al., Program Library, Institut für Kernchemie, Universität Mainz (1975).
- <sup>14)</sup> A. Gilbert and A.G.W. Cameron, Can. J. Phys.  $\underline{43}$ , 1446 (1965).
- 15) F.M. Nuh et al., to be published.
- 16) H. Tovedal and B. Fogelberg, Nucl. Phys. <u>A252</u>, 253 (1975).
- 17) C.M. Jensen et al., Bull. Am. Phys. Soc. <u>21</u> (1976).
- 18) W. Rudolph et al., submitted to J. Inorg. Nucl. Chem
- 19) A. Kerek et al., Nucl. Phys.A195 , 177 (1972).
- 20) E.H. Auerbach and F.G.J. Perey, Brookhaven Natl. Lab. Report BNL 765 (1962).
- 21) E. Lund and G. Rudstam, Proc. Int. Workshop on Gross Properties of Nuclei and Nuclear Excitations III, Hirschegg, Austria (1975) 159.
- <sup>22</sup>) K. Aleklett et al., Nucl. Phys. <u>A246</u>, 425 (1975).
- 23) H.V. Klapdoret al., Verh. Dtsch. Phys. Ges. 1,62 (1974).
- 24) H.V. Klapdor, contribution to this conference.
- 25) A.A. Shihab-Eldin et al., contribution to this conference.