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A Study of the Radiation from Am<sup>241</sup>

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## **A Study of the Radiation from Am<sup>241</sup>**

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With 8 Figures in the Text

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The radiation from the alpha decay of  $\text{Am}^{241}$  has been investigated by studying the fine structure of the alpha spectrum and by performing alpha-gamma and gamma-gamma coincidence measurements. For the daughter nucleus  $\text{Np}^{237}$  a level scheme is proposed which comprises 21 excited states and 39 gamma transitions. Evidence was found for the existence of previously unknown gamma rays at (80), 82, 88, (90), 100, 150 (two transitions), 170, ( $\sim 210$ ), 295, (300), 360, 420, 460 and 690 keV, as well as for the probable occurrence of a new fine structure alpha group at 4754 keV corresponding to a level at 804 keV. For the alpha groups and for most of the gamma rays absolute intensities are given. For some transitions the  $K$ -conversion coefficients and multipolarities were determined. The results are discussed within the framework of the unified model. The level density agrees well with the predictions of the superfluid model and is approximately twice the density yielded from NILSSON's theory.

## 1. Introduction

In the alpha decay of  $\text{Am}^{241}$  various excited states are populated in the daughter nucleus  $\text{Np}^{237}$ . Information on the  $\text{Np}^{237}$  level structure can also be obtained from beta decay of  $\text{U}^{237}$ , electron capture in  $\text{Pu}^{237}$  and from Coulomb excitation experiments. Since the odd-mass nucleus  $\text{Np}^{237}$  is in a region of strongly deformed nuclei, the lower energy levels may be described as being single particle Nilsson-states<sup>1,2</sup>, with rotational levels based upon them<sup>3-5</sup>. More recently, Solov'ev has introduced some modifications in Nilsson's schemes by taking into account the effect of superfluidity<sup>6,7</sup>. Exact knowledge of the excited states of  $\text{Np}^{237}$  thus furnishes a useful contribution to checking the theoretical predictions of these models.

<sup>1</sup> NILSSON, S. G.: Dan. Mat.-Fys. Medd. **29**, No. 16 (1955).

<sup>2</sup> MOTTETSON, B. R., and S. G. NILSSON: Dan. Mat.-Fys. Skr. **1**, No. 8 (1959).

<sup>3</sup> BOHR, A.: Dan. Mat.-Fys. Medd. **26**, No. 14 (1952).

<sup>4</sup> BOHR, A., and B. R. MOTTETSON: Dan. Mat.-Fys. Medd. **27**, No. 16 (1953) and Phys. Rev. **89**, 316 (1953).

<sup>5</sup> BOHR, A., and B. R. MOTTETSON: Beta- und Gamma-Ray Spectroscopy (ed. K. SIEGBAHN), Chapt. XVII. North-Holland Publishing Company: Amsterdam 1955.

<sup>6</sup> SOLOV'EV, V. G.: J. Exptl. Theoret. Phys. (U.S.S.R.) **13**, 456 (1961).

<sup>7</sup> VERESH, T., V. G. SOLOV'EV i T. SHIKLOSH: Bull. Acad. Sci. U.S.S.R., Phys. Ser. **26**, 1053 (1963).

The level structure of Np<sup>237</sup> has been investigated by many authors<sup>8-27</sup>. Level schemes including gamma transitions have been worked out by Day<sup>10</sup>, JAFFE et al.<sup>11</sup>, BARANOV and SHLYAGIN<sup>15</sup>, HOLLANDER et al.<sup>16</sup>, RASMUSSEN et al.<sup>18</sup>, ROSENBLUM et al.<sup>19</sup>, HOFFMANN and DROPEŠKY<sup>21</sup> and SAMOILOV<sup>23</sup>. For the 5/2<sup>+</sup> [642] ground state rotational band (with energy levels at 0, 33 and 77 keV), the 5/2<sup>-</sup> [523] rotational band (with levels at 60, 103, 158 and 225 keV) as well as for the 3/2<sup>-</sup> level at 268 keV, in general, consistency has been achieved among these investigations. RASMUSSEN et al. found evidence for a 1/2<sup>+</sup> [400] rotational band with members at 332, 369 and 371 keV. The results of these authors cast some doubt on the existence of a level at 431 keV reported in earlier papers. GOLDIN et al.<sup>12</sup> observed a low intensity alpha group in the decay of Am<sup>241</sup>, corresponding to a level at 305 keV in Np<sup>237</sup>. No gamma rays or conversion electrons associated with this group could be detected. SAMOILOV attributed a ground state transition to the 305 keV level interpreting this state as the 13/2<sup>-</sup> member of the 5/2<sup>-</sup> [523] rotational band. However, considering the large spin difference to the 5/2<sup>+</sup> ground state, the data are not conclusive. Most recently, BARANOV et

<sup>8</sup> ASARO, F., F. L. REYNOLDS, and I. PERLMAN: Phys. Rev. **87**, 277 (1952).

<sup>9</sup> BELING, J. K., J. O. NEWTON, and B. ROSE: Phys. Rev. **87**, 670 (1952).

<sup>10</sup> DAY, P. P.: Phys. Rev. **97**, 689 (1955).

<sup>11</sup> JAFFE, H., T. O. PASSELL, C. I. BROWNE, and I. PERLMAN: Phys. Rev. **97**, 142 (1955).

<sup>12</sup> GOL'DIN, L. L., E. F. TRET'YAKOV, and G. I. NOVIKOVA: Conf. of the Acad. of Sci. of the U.S.S.R. on the Peaceful Uses of Atomic Energy, July 1-5, 1955, M. (Trans. Consultants Bureau, N. Y., 1955).

<sup>13</sup> TURNER, J. F.: Phil. Mag. **46**, 687 (1955).

<sup>14</sup> NEWTON, J. O.: Nature **175**, 1028 (1955).

<sup>15</sup> BARANOV, S. A., i K. N. SHLYAGIN: J. Exptl. Theoret. Phys. (U.S.S.R.) **3**, 200 (1956).

<sup>16</sup> HOLLANDER, J. M., W. G. SMITH, and J. O. RASMUSSEN: Phys. Rev. **102**, 1372 (1956).

<sup>17</sup> MAGNUSSON, L. B.: Phys. Rev. **107**, 161 (1957).

<sup>18</sup> RASMUSSEN, J. O., F. L. CANAVAN, and J. M. HOLLANDER: Phys. Rev. **107**, 141 (1957).

<sup>19</sup> ROSENBLUM, S., M. VALADARES et J. MILSTED: J. phys. radium **18**, 609 (1957).

<sup>20</sup> BUNKER, M. E., J. P. MIZE, and J. W. STARNER: Bull. Am. Phys. Soc. **2**, 104 (1957).

<sup>21</sup> HOFFMAN, D. C., and B. J. DROPEŠKY: Phys. Rev. **109**, 1282 (1958).

<sup>22</sup> NEWTON, J. O.: Nuclear Phys. **5**, 218 (1958).

<sup>23</sup> SAMOILOV, P. S.: Bull. Akad. Sci. U.S.S.R., Phys. Ser. **23**, 1401 (1959).

<sup>24</sup> BARANOV, S. A., V. M. KULAKOV, V. M. SHATINSKY, i A. G. ZELENKOV: J. Exptl. Theoret. Phys. (U.S.S.R.) **43**, 795 (1962); — Nuclear Phys. **43**, 547 (1963).

<sup>25</sup> BARANOV, S. A., V. M. KULAKOV, and V. M. SHATINSKY: Nuclear Phys. **56**, 252 (1964).

<sup>26</sup> LEDERER, C. M.: UCRL-11028.

<sup>27</sup> WOLFSON, J. L., and J. J. H. PARK: Can. J. Phys. **42**, 1387 (1964).

al.<sup>24, 25</sup>. \* studied the fine structure of the  $\text{Am}^{241}$  alpha spectrum with a high resolution magnetic alpha spectrograph. These measurements revealed several new alpha groups corresponding to levels at 357, 395, 438, 456, 463, 485, (549,) 723 and 758 keV. To the state at 357 keV BARANOV et al. assigned the quantum numbers  $3/2^+$  [651]. Energetically, the levels at 395, 438, 456, 463, 485 and 549 keV fit well into rotational bands belonging to the Nilsson orbitals  $5/2^-$  [523],  $3/2^-$  [521] and  $1/2^+$  [400]. The theoretical interpretation is thus mainly based on energy considerations. Up till now no data existed on the de-excitation of all these levels. For unambiguous classifications such data are very useful. As has been pointed out by Baranov et al., the two levels at 723 and 758 keV possibly belong to the  $\beta$ -vibrational band of the  $5/2^-$  [523] state. Recently, LEDERER<sup>26</sup> performed alpha-gamma and alpha-conversion electron coincidence measurements in this energy region. His results support the interpretation given by BARANOV et al.

The present study was undertaken to refine and amplify the data on the levels and gamma transitions occurring in  $\text{Np}^{237}$ . Significant experimental difficulties arise from the fact that alpha groups feeding levels higher than 158 keV are very weak, the intensity being only  $10^{-2}$  to  $10^{-4}$  %. The primary tool was a recently developed apparatus for measuring alpha-gamma coincidences.

## 2. Experimental Procedure

The  $\text{Np}^{237}$  level structure was investigated by studying the fine structure of the  $\text{Am}^{241}$  alpha spectrum and by performing alpha-gamma and gamma-gamma coincidence measurements.

In the alpha-gamma coincidence spectrometer a 25 mm<sup>2</sup> silicon surface barrier detector was used for detection of the alpha particles. The depletion depth was about 100  $\mu$  at 50 V bias. The total resolution including amplifier noise, energy loss in the source and long-term instabilities in the electronic circuit was about 25 keV. The gamma detector consisted of a  $1\frac{1}{2}'' \text{ } \varnothing \times 1''$  NaI(Tl) crystal mounted on an EMI 6097 A photomultiplier. For the  $\text{Cs}^{137}$  gamma ray the energy resolution was 7.4%. The effective resolving time of the coincidence circuit was selected to 0.5  $\mu$ sec. Only pulses corresponding to gamma ray energies  $> 60$  keV were permitted to trigger the coincidence. Pile-up effects due to the intense 60 keV radiation were reduced by an absorber of 0.5 mm cadmium between source and gamma ray detector. Data were taken in several runs using a 1024 channel two-dimensional pulse height analyser.

\* The more extensive second paper appeared after completion of the fine structure measurements described in section 3.1. The results are in excellent agreement.

In directly determined alpha spectra the weak groups are masked by the low-energy tails of the intense peaks from the main transitions. Alpha fine structure measurements were, therefore, performed by using the alpha-gamma coincidence circuit. Since the decay to higher excited states occurs in coincidence with high-energy gamma rays, the low-energy tails of the intense peaks can be eliminated by means of an integral discriminator in the gamma pulse channel. Employing this method even alpha groups with an intensity of  $10^{-4}$  % or below were clearly observed.

For gamma-gamma coincidence measurements two identical detectors were used consisting of  $1\frac{1}{2}'' \text{ } \varnothing \times 2''$  NaI(Tl) crystals mounted on RCA 6810 A photomultipliers. The resolution was 8.1% and 8.4%, respectively, for the Cs<sup>137</sup> gamma ray. A Compton shield of 6 mm thick lead surrounded by 0.5 mm cadmium and 0.5 mm copper was placed between the detectors in order to avoid counter-to-counter scattering. The Bell-type fast coincidence circuit had an effective resolving time of about 60 nsec. Data were analysed in  $256 \times 256$  channels by means of a  $2 \times 10$  bit dual ADC. The digital data were processed and stored via one of the input channels of the CDC 160 A on-line computer installed<sup>28</sup> at the Karlsruhe research reactor. The source strength for the gamma ray studies was 106  $\mu\text{C}$ . In order to reduce the counting rate due to the intense 60 keV radiation cadmium absorbers with up to 1 mm thickness were used.

Alpha sources were prepared by drying up a 1 n HCl solution containing 12  $\mu\text{g/ml}$  Am<sup>241</sup> onto a 0.2 mm fire-proof glass backing. These samples were heated in order to get an americium oxide layer as pure and uniform as possible. The source strength was about 2  $\mu\text{C}$ .

### 3. Experimental Results

**3.1. Fine Structure of the Alpha Spectrum.** The directly measured alpha spectrum only reveals the well-known alpha groups at 5389, 5443, 5486, 5513 and 5545 keV. The weak 5469 keV group<sup>24, 25</sup>, corresponding to a transition to the 77 keV level, is obscured by the closely neighbouring most intense alpha group.

Using the coincidence method the fine structure of the alpha spectrum is clearly observed. In Fig. 1 the results are shown for three different discriminator settings in the gamma ray channel. The spectra were analysed employing the method proposed by PREUSS and ESCARFAIL<sup>29</sup>. Some weak peaks in the spectra are attributed to small isotopic impurities present in the source. Comparison with the magnetic spectrograph

<sup>28</sup> KRÜGER, G., and G. DIMMLER: Automatic Acquisition and Reduction of Nuclear Data, Proc. of an EANDC-Conf. (ed. K. H. BECKURTS, W. GLÄSER and G. KRÜGER), p. 149.

<sup>29</sup> PREUSS, L. G., and J. P. ESCARFAIL: Nuclear Instr. and Meth. 9, 212 (1960).

measurements performed by BARANOV et al. (cf. section I) reveals excellent agreement regarding both the existence of the new alpha groups and

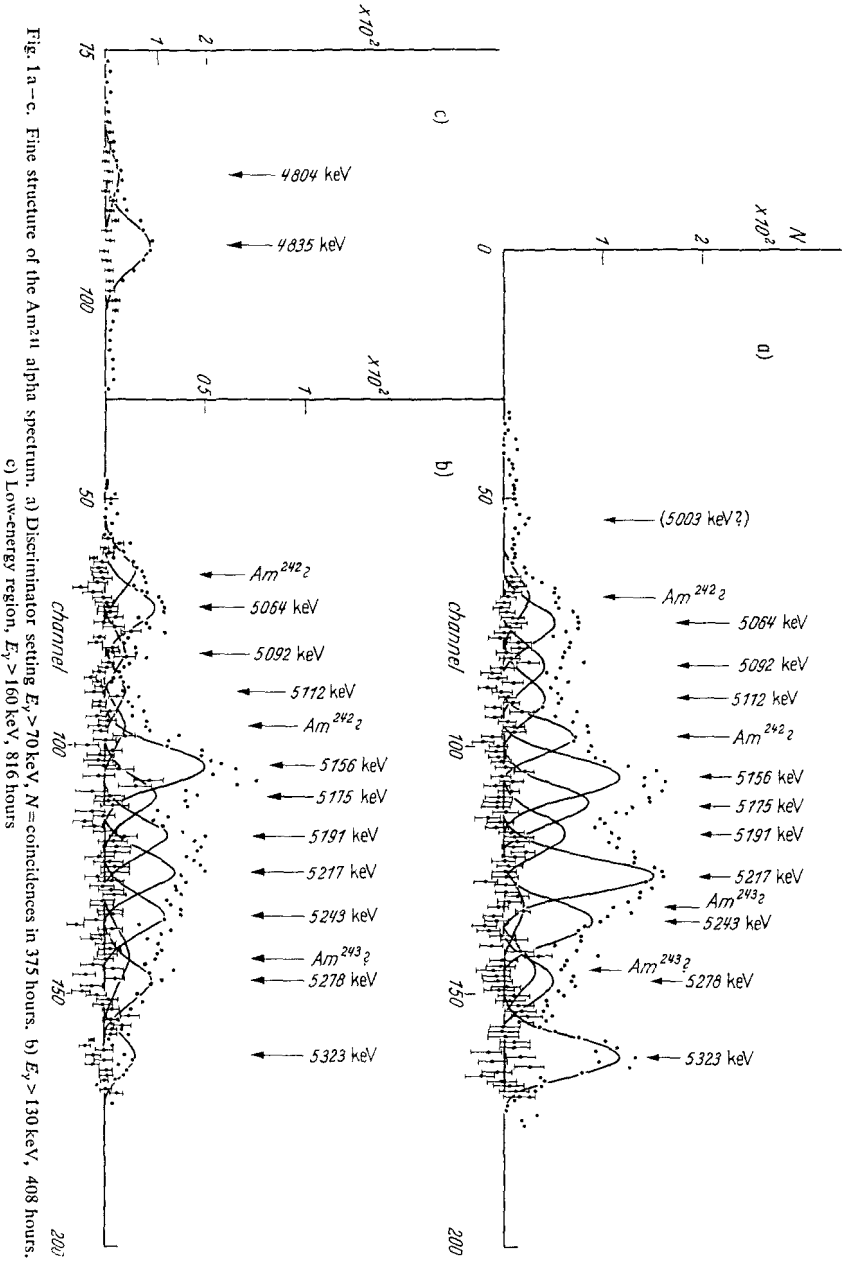


Fig. 1a-c. Fine structure of the  $Am^{241}$  alpha spectrum. a) Discriminator setting  $E_d > 70$  keV,  $N =$  coincidences in 375 hours. b)  $E_d > 130$  keV, 408 hours. c) Low-energy region,  $E_d > 160$  keV, 816 hours.

the energy values. The doublets observed by these authors at 5089/5096 keV and 5178/5182 keV remain unresolved in Fig. 1 and appear as peaks with energies at 5092 and 5175 keV.

**3.2. Gamma Ray Spectrum.** The total gamma ray spectrum was investigated by means of one of the  $1\frac{1}{2}'' \text{ } \varnothing \times 2''$  NaI(Tl) crystals using

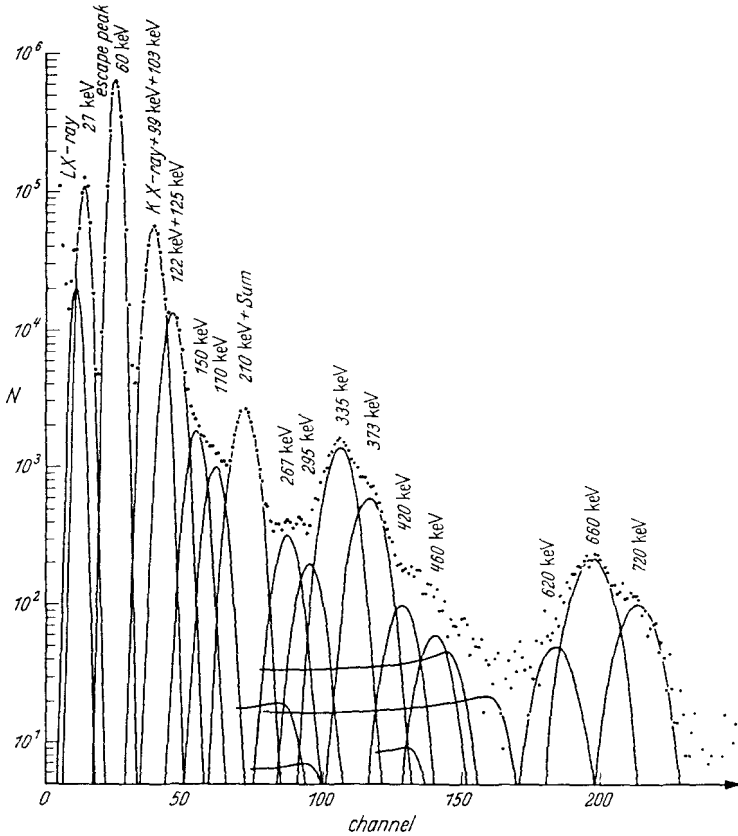


Fig. 2. Am<sup>241</sup> gamma ray spectrum taken with a  $1\frac{1}{2}'' \text{ } \varnothing \times 2''$  NaI(Tl) crystal; absorber 1 mm cadmium

cadmium absorbers of different thicknesses. With 1 mm cadmium the number of counts per channel could be displayed over five orders of magnitude for the whole energy range. This spectrum is shown in Fig. 2. Special attention was paid to the background subtraction. In order to reduce the background counting rate the spectra were taken in a large 10 cm thick lead box. Again, the spectra were analysed by the method of PREUSS and ESCARFAIL. In addition, for the high-energy transitions the Compton distribution was taken into account. The data were deduced from known spectrum shapes.



Analysis of the total gamma ray spectrum revealed the existence of new gamma transitions with the following energies: 150, 295, 420 and 460 keV.

**3.3. Gamma-Gamma Coincidence Measurements.** In these measurements data were taken using 1 mm cadmium absorbers for both detectors. Again, the background was reduced by placing the counters in a large lead box. The dual ADC was adjusted to record coincidences over the whole energy range of the gamma ray spectrum. However, pronounced peaks were observed only in the low-energy region. This indicates that the high-energy transitions ( $E_\gamma > 210$  keV) either reach the ground state or the levels at 33, 60 and 77 keV. Transitions de-exciting these levels are highly converted and the unconverted photons are strongly attenuated by 1 mm cadmium.

Table 1. *Summary of the gamma-gamma coincidence measurements*

Gamma energy range (keV)	Coincident gamma ray energies (keV)						
	60	K 99 100 103	122 125	150	160	170	210
40–70	+	+	+	+		(+)	(+)
80–135	+	+	+	(+)		+	+
135–165		+	+	(+)			+
170–195		+	+	+			+
200–250		+			(+)		+

The most significant spectra are shown in Fig. 3. The results are summarized in Table 1. Along with previously known cascades the following new coincidence relationships can be deduced from the gamma-gamma coincidence data: 60 keV–150 keV, (99 keV–150 keV), 122 keV–170 keV, 125 keV–150 keV, 150 keV–170 keV, ( $\sim 100$  keV–210 keV) and  $\sim 210$  keV–210 keV. These conclusions are supported by the alpha-gamma coincidence measurements described in subsection 3.4. The occurrence of coincidences between the 210 keV transition and gamma rays of about 160 keV confirms the results reported by DAY<sup>10</sup>, BARANOV and SHLYAGIN<sup>15</sup> and HOLLANDER et al.<sup>16</sup>.

**3.4. Alpha-Gamma Coincidence Measurements.** In the present work main information on the decay of Am<sup>241</sup> was obtained from alpha-gamma coincidence studies. The spectra of gamma rays coincident with alpha groups corresponding to levels with energies  $\geq 158$  keV are shown in Figs. 4–6 and will be briefly discussed here. We premise that due to the high level density some spectra contain contributions from neighbouring alpha groups. The spectra are not corrected for random coincidences. Because of the long run time these coincidences were recorded

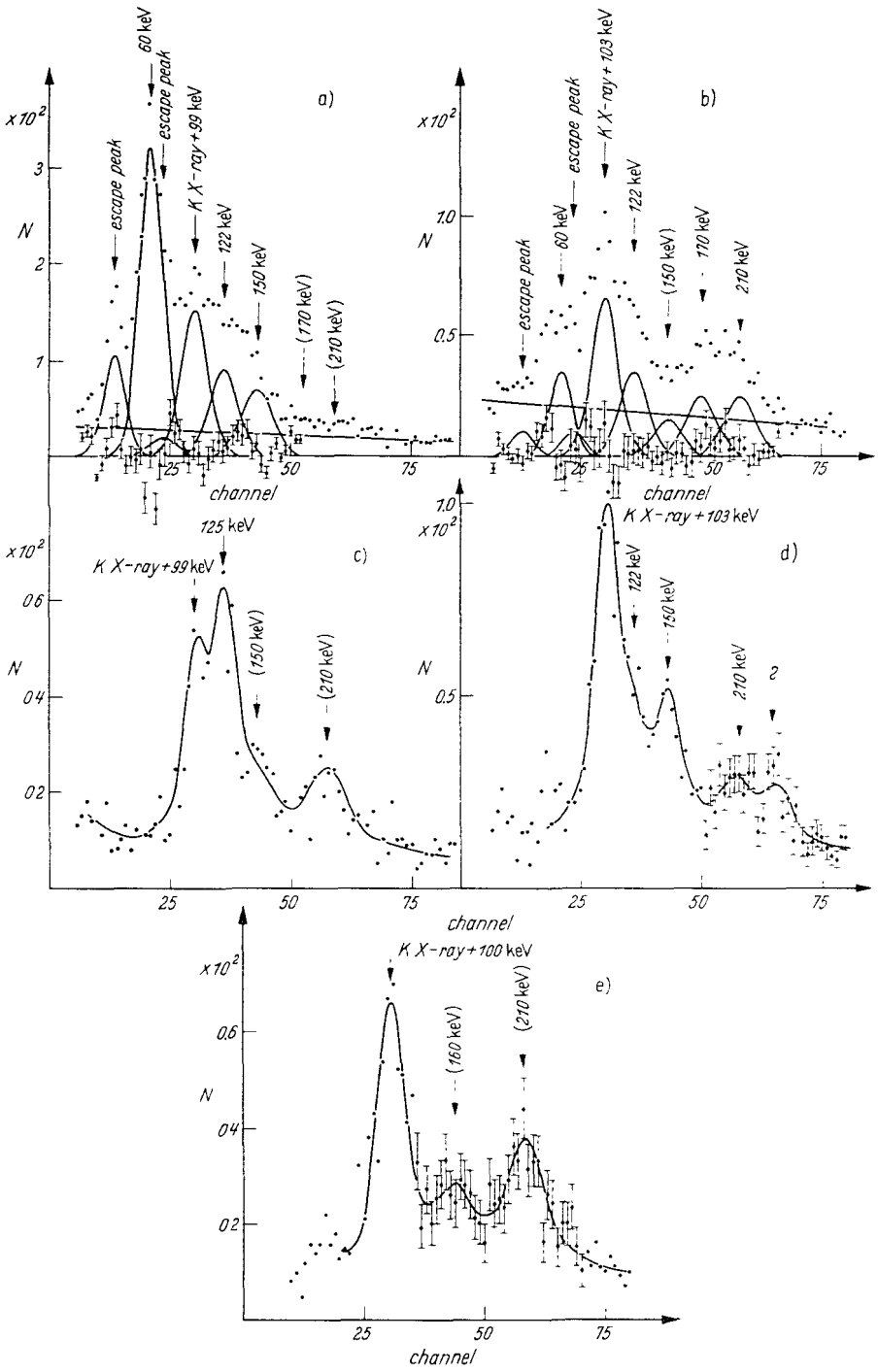


Fig 3a—e. Spectrum of gamma rays coincident with the photon energy range from a) 40 keV to 70 keV, b) 80 keV to 135 keV, c) 135 keV to 165 keV, d) 170 keV to 195 keV, e) 200 keV to 250 keV. 1 mm cadmium absorbers for both detectors

with poorer resolution. Noticeable contributions were observed only for the photopeak at 100 keV. Where information was needed from this peak appropriate corrections were made.

As can be seen from Fig. 4a, the level at 159 keV is partly de-excited by a 125 keV transition to the  $7/2^+$  member of the ground state rotational band. This result is in agreement with the suggestion of several other investigations<sup>10, 11, 23</sup>. In the gamma ray spectrum corresponding to the 225 keV state (Fig. 4b), along with previously known transitions<sup>15, 23</sup>, a weak line is observed at 150 keV. This indicates that the 225 keV level is also de-excited in part by a transition to the ground state rotational band. The spectrum shown in Fig. 4c confirms the results of previous authors<sup>10, 11, 15, 16, 18, 23</sup> regarding the pattern of transitions from the level at 271 keV.

The alpha group appearing at 5243 keV in the fine structure measurements corresponds to a level at 307 keV (cf. section 1). This energy fits well into the  $5/2^-$  [523] rotational band. Using the formula of BOHR and MOTTELSON the  $13/2^-$  member of this band is predicted at 306 keV. The alpha-gamma coincidence shown in Fig. 4d clearly indicates that the classification  $13/2^-$   $5/2^-$  [523] is correct. Considering the coincidence relationships deduced in subsection 3.3 and the intensity ratios of the 122 keV and 150 keV gamma rays in Figs. 4b and 4d, the 150 keV transitions observed in these spectra are certainly not identical. The 150 keV gamma ray appearing in coincidence with the 5243 keV alpha group is interpreted as being the  $E2$  transition which is expected between the  $13/2^-$  and  $9/2^-$  rotational members at 307 and 158 keV. This conclusion then immediately reveals the relationships 60 keV—150 keV, 99 keV—150 keV and 125 keV—150 keV observed in the gamma-gamma coincidence studies. According to the well-known branching of transitions from the 158 keV level the 125 keV gamma ray is not strong enough to explain the peak at this energy in Fig. 4d. Therefore, we are forced to conclude that this peak corresponds to the  $E2$  transition between the levels at 225 and 103 keV. The occurrence of the 122 keV gamma ray in coincidence with the 5243 keV alpha group implies the existence of a new 82 keV transition going from the 307 keV level to the level at 225 keV. A ground state transition, as reported by SAMOILOV<sup>23</sup>, could not be verified.

The gamma ray spectrum coincident with the 5217 keV alpha group (Fig. 4e) illustrates that the 333 keV level is predominantly de-excited through the level at 271 keV. The ground state transition is much weaker than is expected from the gamma singles spectrum. Hence the photopeak at 335 keV in Fig. 2 must be due to a doublet. This result is in excellent agreement with the data obtained by RASMUSSEN et al.<sup>18</sup> from the beta decay of U<sup>237</sup>.

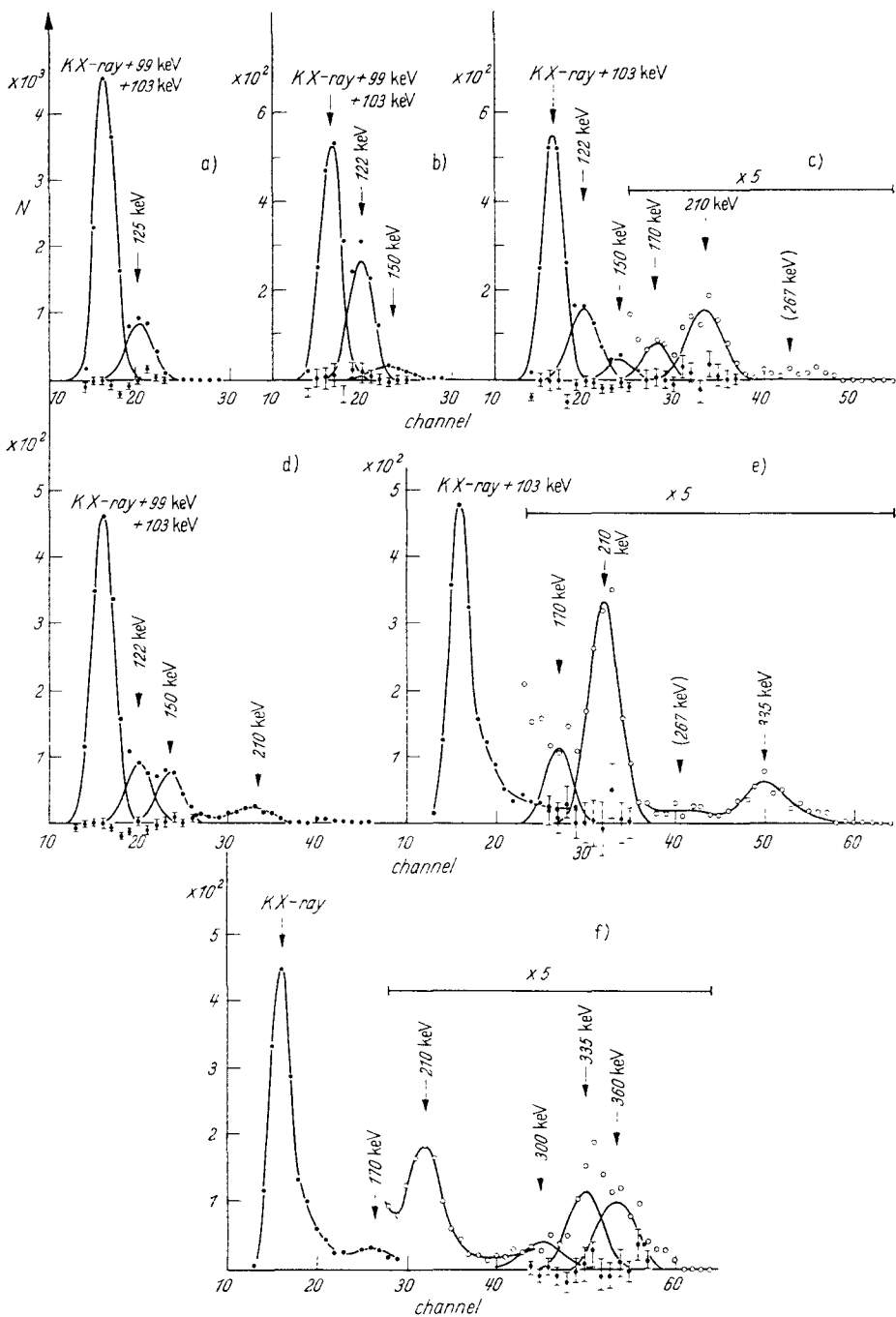


Fig. 4a–f. Alpha-gamma coincidence spectra. a) Alpha energy range 5369–5402 keV. b) 5305–5336 keV. c) 5257–5297 keV. d) 5236–5255 keV. e) 5207–5227 keV. f) 5183–5203 keV. a)–c) Coincidences in 355 hours; energy scale 6.5 keV per channel. d)–f) Coincidences in 450 hours; 6.9 keV per channel

The 5191 keV fine structure alpha group implies a level at 360 keV. The corresponding gamma ray spectrum is shown in Fig. 4f. Analysis of the spectrum revealed a new gamma ray with an energy of 360 keV. The photopeaks at 210 and 335 keV are mainly attributed to interference

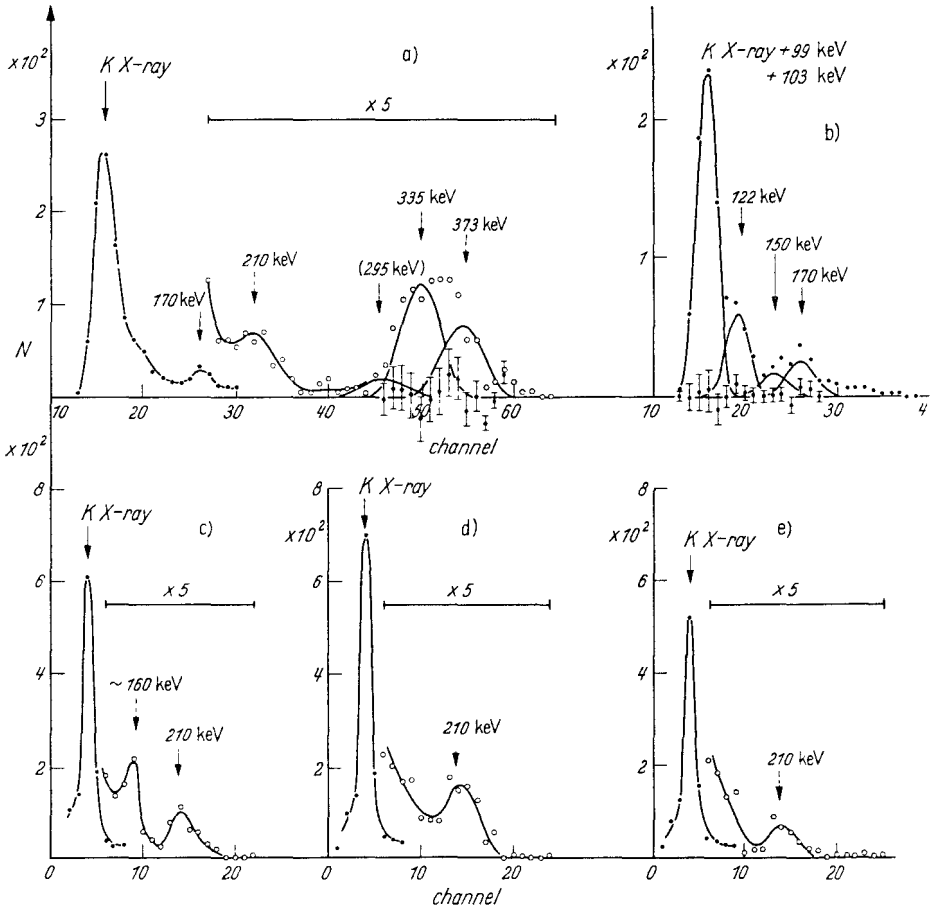


Fig. 5a-e. Alpha-gamma coincidence spectra. a) Alpha energy range 5169-5183 keV. b) 5151-5165 keV. c) 5110-5122 keV. d) 5084-5096 keV. e) 5056-5068 keV. a)-b) Coincidences in 450 hours, energy scale 6.9 keV per channel. c)-e) Coincidences in 690 hours; 11 keV per channel; low energy region

from neighbouring alpha groups. Probably, the 360 keV peak contains some counts from the 373 keV gamma ray. The measurement may indicate that a weak transition of about 300 keV feeds the level at 60 keV.

The gamma ray spectrum corresponding to the level doublet at 369 and 373 keV is displayed in Fig. 5a. Gamma rays are seen at 210, (295), 335 and 373 keV. Considering the spectrum shapes in Figs. 4e and 5a, we conclude that the 335 keV transition is different from that appearing

in Fig.4e. In agreement with RASMUSSEN et al.<sup>18</sup> it is interpreted as proceeding to the level at 33 keV. The 295 keV gamma ray (cf. Fig.2) fits to the energy difference between the 369 keV level and the  $9/2^+$  member of the ground state rotational band. The photopeak at 210 keV is stronger than is expected from interference of neighbouring alpha groups. This indicates the existence of a weak transition to the 271 keV state. The probable occurrence of the cascade relationship  $\sim 100$  keV – 210 keV observed in the gamma-gamma coincidence measurements (cf. subsection 3.3) is in favour of this conclusion. RASMUSSEN et al.<sup>18</sup> reported the possible existence of a transition reaching the 333 keV state. On the basis of the present experiments such a transition cannot be ruled out.

The alpha group appearing at 5156 keV in the fine structure measurements implies a level at 395 keV. The  $15/2^-$  member of the  $5/2^-$  [523] rotational band is predicted to occur at about 392 keV. As can be seen from the alpha-gamma coincidence spectrum shown in Fig.5b, it is correct to assign the 395 keV level as  $15/2^-$   $5/2^-$  [523]. The previously unknown 170 keV transition is interpreted as being the  $E2$  transition proceeding to the  $11/2^-$  rotational level at 225 keV. The presence of the 122 keV gamma ray and the occurrence of the cascade relationships 122 keV – 170 keV and 150 keV – 170 keV cited in subsection 3.3 are then easily explained. As a result of intensity considerations the 150 keV transition observed in coincidence with the 5323 keV alpha group can be ruled out for interpretation of the 150 keV peak in Fig.5b. Obviously, this peak corresponds to the  $E2$  transition going from the 307 keV level to the  $9/2^-$  level at 158 keV. Hence, a new 88 keV transition must be postulated feeding the 307 keV state.

A level at 440 keV results from the fine structure alpha group at 5112 keV. The alpha-gamma coincidence spectrum (Fig.5c) suggests that this level is mainly de-excited through the state at 271 keV (cf. subsection 3.3). The possible occurrence of a weak line at 360 keV in the high-energy spectrum may indicate that a weak transition of about 80 keV feeds the level at 360 keV. The existence of a weak ground state transition cannot be ruled out.

The alpha groups at 5096 and 5089 keV correspond to energy levels at 456 and 463 keV, respectively. The new gamma rays observed at 420 and 460 keV (cf. subsection 3.2) are attributed to these levels. In the alpha-gamma coincidence spectrum photopeaks at 420 and 460 keV possibly occur. However, due to the poor counting rate the coincidence relationships could not be definitely established. The presence of 210 keV gamma rays in the low-energy spectrum (Fig. 5d) is attributed to a partial de-excitation of these levels to the doublet at 370 keV.

The alpha group appearing at 5064 keV in the fine structure measurements implies a level at 489 keV. The gamma ray spectrum is shown in Fig. 5e. The occurrence of a peak at 210 keV may be explained by a transition to the 271 keV state. This conclusion is supported by the  $\sim 210-210$  keV cascade relationship observed in the gamma-gamma coincidence measurements.

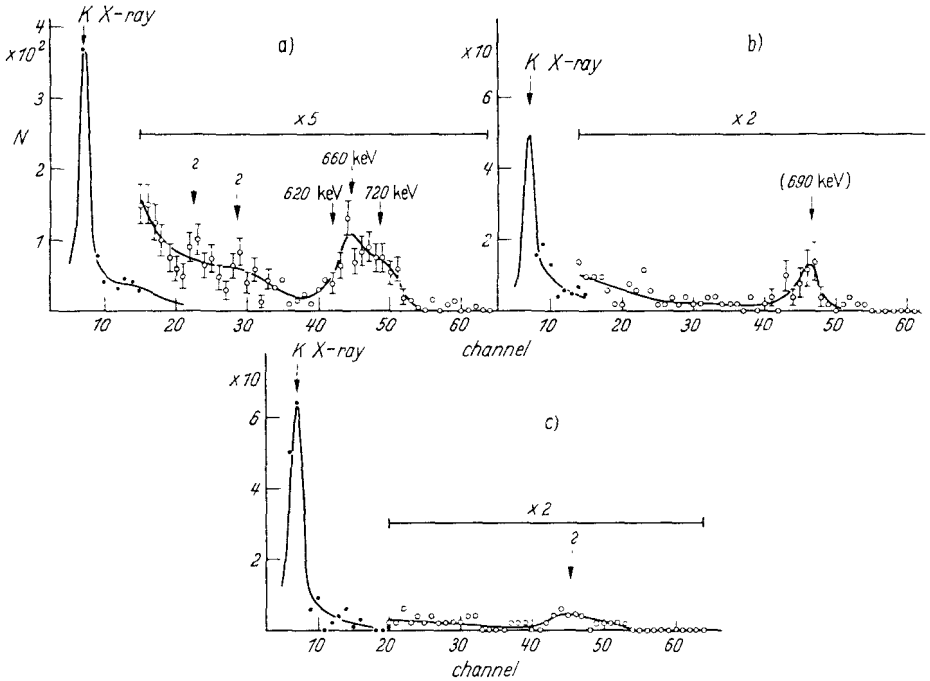


Fig. 6 a - c. Alpha-gamma coincidence spectra. Coincidences in 545 hours; energy scale 15 keV per channel. a) Alpha energy range 4823 - 4843 keV. b) 4793 - 4813 keV. c) 4743 - 4763 keV

The spectrum of gamma rays de-exciting the level at 722 keV is displayed in Fig. 6a. Excellent agreement is found with the data reported by LEDERER<sup>26</sup>. An additional transition proceeding from the 722 keV level was seen by this author at 698 keV. The present experiments are not in contrast to the existence of such a transition.

The alpha-gamma coincidence spectrum corresponding to the energy level at 753 keV reveals the probable existence of a new gamma ray at 690 keV (Fig. 6b) which is interpreted as going from the 753 keV level to the intrinsic state at 60 keV.

The integral number of counts in the high-energy region showed an additional maximum at an alpha energy of about 4754 keV. This may indicate the presence of a new very weak alpha group revealing a level

at 804 keV. Energetically, this level fits well into the  $\beta$ -vibrational band. The poor counting rate precludes a statement on the gamma rays de-exciting this level (Fig. 6c).

The results of the alpha-gamma coincidence measurements are summarized in Table 2.

Table 2. Summary of the alpha-gamma coincidence measurements

Alpha energy range (keV)	Coincident gamma ray energies (keV)																	
	K 99 103	122 125	150	160	170	210	267	295 300	335	360	373	420	460	620	660	690	720	
5369-5402	+	+																
5305-5336	+	+	+															
5257-5297	+	+	+		+	+	(+)											
5236-5255	+	+	+				(+)											
5207-5227	+				+	+	(+)		+									
5183-5203	+				+	+		(+)	+	+								
5169-5183	+				+	+		(+)	+			+						
5151-5165	+	+	+		+													
5110-5122	+			+		+						(+)						
5084-5096	+					+			(+)		(+)	(+)	(+)	(+)				
5056-5068	+					+												
4823-4843	+														+	+		+
4793-4813	+																(+)	
4743-4763	+																	

**3.5. Gamma Ray Energies and Intensities. Errors.** Table 3 gives the unconverted gamma ray intensities which could be deduced from the present experiments. Most of the values were obtained from the gamma singles spectrum. In some cases alpha-gamma coincidence spectra were used. For evaluating intensities from these measurements the spectra had first to be corrected for contributions from neighbouring alpha groups. All values given in Table 3 are corrected for crystal efficiency, peak-to-total ratio and absorption between source and detector. The errors assigned to the intensities of unconverted photons include statistical errors, estimates of errors introduced in treating incompletely resolved lines and estimates of errors introduced in determining the contribution from the tails of nearby alpha groups. In general, the uncertainties in the corrections for crystal efficiency, peak-to-total ratio and absorption were negligible.

The maximum estimated errors for the gamma ray energies have been included in column 1 of Table 3.



Table 3. *Gamma ray intensities*

$E_\gamma$ (keV)	Relative intensity of unconverted photons	Estimated error (%)	Absolute intensity of unconverted photons (%)
60	100	15	38
99	$5.2 \times 10^{-2}$	30	$2.0 \times 10^{-2}$
103	$4.0 \times 10^{-2}$	30	$1.5 \times 10^{-2}$
$122 \pm 2$	$3.3 \times 10^{-3}$	25	$1.3 \times 10^{-3}$
$125 \pm 2$	$1.0 \times 10^{-2}$	25	$3.8 \times 10^{-3}$
$150 \pm 5 (\rightarrow 9/2^+)$	$2.9 \times 10^{-4}$	35	$1.1 \times 10^{-4}$
$150 \pm 5 (\rightarrow 9/2^-)$	$5.6 \times 10^{-4}$	35	$2.1 \times 10^{-4}$
$160 \pm 8$	$8.2 \times 10^{-5}$	50	$3.1 \times 10^{-5}$
$170 \pm 5 (\rightarrow 7/2^-)$	$3.6 \times 10^{-4}$	35	$1.4 \times 10^{-4}$
$170 \pm 5 (\rightarrow 11/2^-)$	$1.2 \times 10^{-4}$	35	$4.6 \times 10^{-5}$
$210 \pm 2 (\rightarrow 5/2^-)$	$1.5 \times 10^{-3}$	20	$5.7 \times 10^{-4}$
$210 \pm 10 (\rightarrow 3/2^-)$	$< 1 \times 10^{-4}$	—	$< 4 \times 10^{-5}$
$267 \pm 6$	$2.4 \times 10^{-4}$	25	$9.1 \times 10^{-5}$
$295 \pm 6^*$	$1.7 \times 10^{-4}$	30	$6.5 \times 10^{-5}$
$335 \pm 7 (\rightarrow 5/2^+)$	$2.9 \times 10^{-4}$	20	$1.1 \times 10^{-4}$
$335 \pm 7 (\rightarrow 7/2^+)$	$1.2 \times 10^{-3}$	20	$4.6 \times 10^{-4}$
$360 \pm 10$	$\sim 5 \times 10^{-4}$	—	$\sim 2 \times 10^{-4}$
$373 \pm 7$	$7.5 \times 10^{-4}$	25	$2.9 \times 10^{-4}$
$420 \pm 10$	$1.6 \times 10^{-4}$	30	$6.1 \times 10^{-5}$
$460 \pm 10$	$1.1 \times 10^{-4}$	30	$4.2 \times 10^{-5}$
$620 \pm 10$	$2.0 \times 10^{-4}$	25	$7.6 \times 10^{-5}$
$660 \pm 6$	$8.7 \times 10^{-4}$	20	$3.3 \times 10^{-4}$
$690 \pm 20$	$1.9 \times 10^{-4}$	30	$7.2 \times 10^{-5}$
$720 \pm 7$	$4.5 \times 10^{-4}$	20	$1.7 \times 10^{-4}$

**3.6. Alpha Energies and Intensities. Errors.** For most of the alpha groups the maximum error of the energies is estimated to be  $\pm 4$  keV, that of the alpha energies at 4754, 4804 and 5003 keV is estimated to be  $\pm 8$  keV. The same errors apply to the corresponding level energies. The most intense  $\text{Am}^{241}$  alpha group was taken as the energy standard. The absolute energy of this group has been measured by LEANG<sup>30</sup> to be  $5486.0 \pm 0.9$  keV. According to a recent recalibration of alpha particle energies<sup>31</sup> the values reported in the present paper may be shifted by  $+1.5$  keV. The level energies, however, remain unaffected. Table 4 summarizes the results on the fine structure of the alpha spectrum. The energy values at 5089, 5096, 5178, 1582, 5469, 5513 and 5545 keV were adopted from Ref. <sup>25</sup>. Alpha intensities were calculated from the gamma ray intensities assuming reasonable multipolarities and employing

\* Including the possible 300 keV gamma ray.

<sup>30</sup> LEANG, C. F.: *Compt. rend.* **255**, 3155 (1962).

<sup>31</sup> WAPSTRA, A. H.: *Nuclear Phys.* **57**, 48 (1964).

experimental or theoretical *K*- and *L*-conversion coefficients<sup>32-35</sup>. The intensities for the alpha groups at 5389, 5443, 5486, 5513 and 5545 keV were determined from the directly measured alpha spectrum. Analysis was performed by presuming gaussian line shapes with appropriate low-energy tails characteristic for semiconductor detectors. As the 5469 keV alpha group is obscured by the intense 5486 keV radiation, only an upper limit for the intensity of this group can be given. The results listed in Table 4 are in fair agreement with the data reported by BARANOV et al.<sup>24,25</sup>.

Table 4. *Fine structure of Am<sup>241</sup> alpha spectrum*

Alpha energy (keV)	Intensity (%)	Hindrance factor <i>F</i>	Alpha energy (keV)	Intensity (%)	Hindrance factor <i>F</i>
5545	0.35 ± 0.08	690	5182	(1.2 ± 0.3) × 10 <sup>-3</sup>	
5513	0.21 ± 0.05	750	5178		
5486	85.6 ± 1.0	1.3	5156	(5.0 ± 1.8) × 10 <sup>-4</sup>	2400
5469	< 0.1	> 890	5112	(3.5 ± 2.1) × 10 <sup>-4</sup>	2000
5443	12.3 ± 0.6	5.1	5096	} ~ 5 × 10 <sup>-4</sup>	
5389	1.5 ± 0.3	20	5089		
5323	(1.4 ± 0.4) × 10 <sup>-2</sup>	890	5064	< 2 × 10 <sup>-4</sup>	> 1600
5278	(4.1 ± 2.0) × 10 <sup>-4</sup>	16000	5003	< 2 × 10 <sup>-4</sup>	> 600
5243	(2.8 ± 0.9) × 10 <sup>-3</sup>	1500	4835	(6.5 ± 1.3) × 10 <sup>-4</sup>	14
5217	(1.1 ± 0.3) × 10 <sup>-3</sup>	2600	4804	(8.4 ± 2.5) × 10 <sup>-5</sup>	67
5191	~ 6 × 10 <sup>-4</sup>	3300	4754	< 1 × 10 <sup>-5</sup>	> 260

**3.7. *K*-Conversion Coefficients. Multipolarities.** For some of the electromagnetic transitions the *K*-conversion coefficients could be determined

Table 5. *K-conversion coefficients. Multipolarities*

Gamma ray energy (keV)	<i>K</i> -conversion coefficient	Multipolarity	Gamma ray energy (keV)	<i>K</i> -conversion coefficient	Multipolarity
210	2.5 ± 0.8	<i>M</i> 1	660	0.3 ± 0.2 <sup>b)</sup>	<i>M</i> 1 <sup>b)</sup>
335(→7/2 <sup>+</sup> )	} 0.51 ± 0.26	a)	690	0.1 ± 0.1	<i>M</i> 1
373			0.1 ± 0.06		

a) If both transitions are *M*1, the total theoretical *K*-conversion coefficient is 0.63 using the intensities given in table 3 and taking into account screening effects and the influence of the finite size of the nucleus.

b) Assuming *E*1 radiation for the 720 keV transition. The data are thus mutually consistent.

<sup>32</sup> ROSE, M. E., H. G. GOERTZEL, and C. L. PERRY: *K*-shell Internal Conversion Coefficient Revised Tables, ORNL 1023.

<sup>33</sup> SLIV, L. A., and I. M. BAND: Coefficients of Internal Conversion of Gamma-Radiation, Part 2, *L*-shell, Translation in NPTR 217 (1958).

<sup>34</sup> SLIV, L. A., and I. M. BAND: *J. Exptl. Theoret. Phys. (U.S.S.R.)* 4, 133 (1957).

<sup>35</sup> SPINRAD, B. I.: *Phys. Rev.* 98, 1302 (1955).

from the alpha-gamma coincidence measurements. The spectra were carefully corrected for random coincidences and for contributions from neighbouring alpha groups. The results, along with the apparent multipolarities, are given in Table 5.

#### 4. Discussion

**4.1.  $\text{Np}^{237}$  Level Scheme.** The level scheme for the nucleus  $\text{Np}^{237}$  based on the present results is shown in Fig.7. It comprises 21 excited states and 39 gamma transitions. There are no reports in the literature regarding previous observation of a level at 804 keV and of gamma rays at 80, 82, 88, 90, 100, 150 (two transitions), 170, 210, 295, 300, 360, 420, 460 and 690 keV\*. The level diagram has been divided into

Table 6.  
*Rotational spacing factors in  $\text{Np}^{237}$*

Rotational band	$\frac{\hbar^2}{2\theta}$ (keV)	$E(2)$ (keV)	Decoupling parameter (keV)
$5/2^+$ [642]	4.75	0.003	—
$5/2^-$ [523]	6.27		—
$1/2^+$ [400]	6.37		+1.10
$3/2^-$ [521]	9.4		—
$\beta$ -vibr.	4.7		—

two parts, the first one containing transitions between levels belonging to different rotational bands, the second one comprising transitions within rotational bands. The investigation both of alpha-gamma and gamma-gamma coincidence relationships provides convincing evidence of the correctness of the proposed decay scheme.

Six Nilsson states and one  $\beta$ -vibration are identified. The spacing factors for five of the corresponding rotational bands are compiled in Table 6.

In the following paragraphs the level structure will be briefly discussed within the framework of the unified model paying special attention to the results of the present study. As regards the ground state rotational band and the well-established low-energy transitions we refer to the extensive papers of previous authors.

**4.2. The Rotational Band Based on the Intrinsic Level at 60 keV.** The level at 60 keV has to be identified with the configuration  $^2 5/2^-$  [523]. As is confirmed in Section 3, the rotational band on this state is developed up to a spin value of  $15/2^-$ . Obviously, the new gamma rays observed at 82, 88, 150 and 170 keV correspond to the  $M 1$  and  $E 2$  transitions

\* *Note added in proof.* Meanwhile we got knowledge of a paper by KAMOUN et al. (Compt. rend. Congrès International de Physique Nucléaire, Paris 1964, Vol. II), in which the existence of gamma transitions at  $162 \pm 5$  and  $355 \pm 10$  keV is reported. Most probably, the 355 keV gamma ray is identical with the transition observed at  $360 \pm 10$  keV in the present study. The 162 keV line may correspond to the 150 keV ( $\rightarrow 9/2^-$ ) transition though the energy value is well outside experimental error. The intensities given by KAMOUN et al. differ considerably from the values quoted in Table 3.

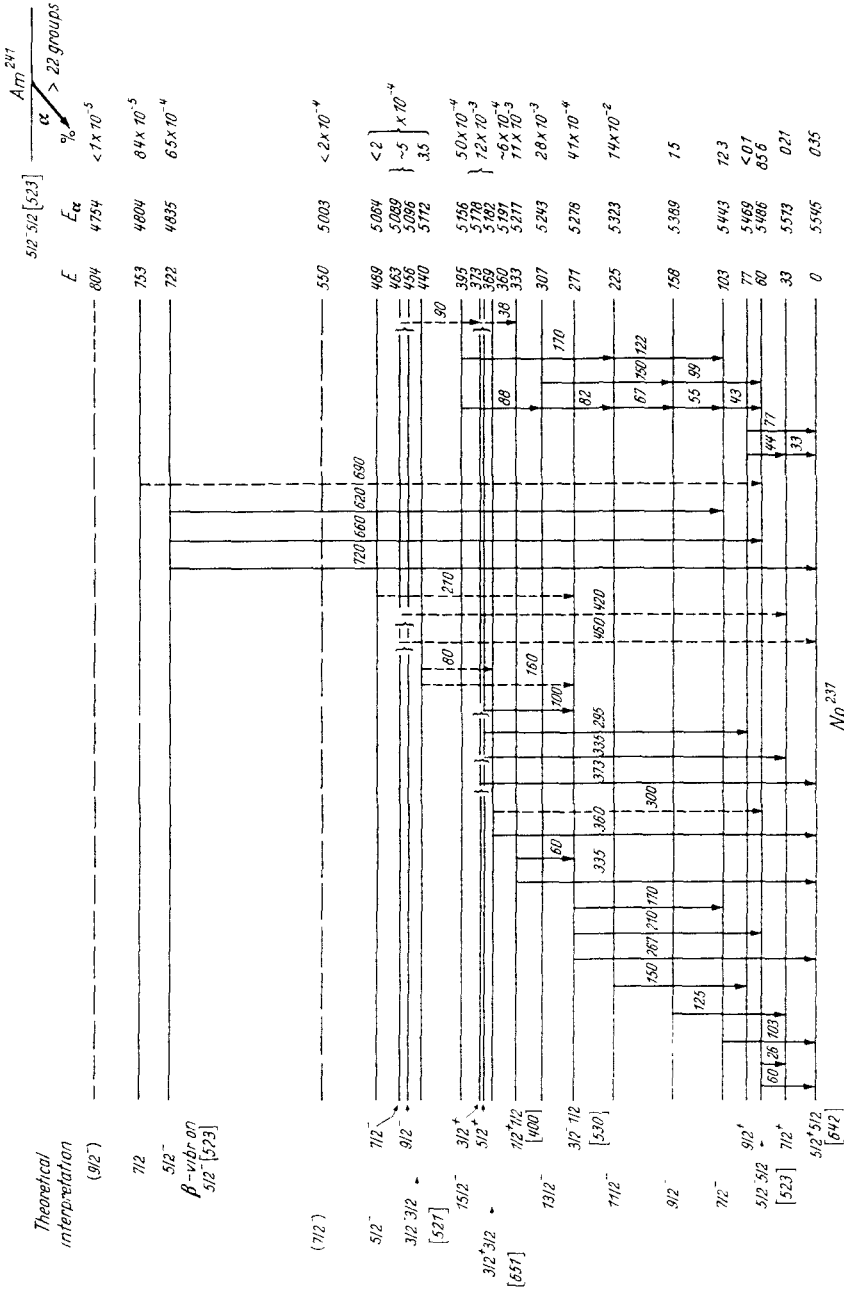


Fig. 7. Level scheme of Np<sup>237</sup> based on the results of the present investigation

between the higher members of this band. Ample evidence was found in Section 3 that the 225 keV level is partly de-excited by an E 1 transition

to the  $9/2^+$  member of the ground state rotational band. The weakness of this transition is explained by a strong hindrance due to the selection rules in terms of the asymptotic quantum numbers.

The levels of the  $5/2^-$  [523] band are excited in the so-called favoured alpha transitions. The intensity ratios of favoured transitions were calculated from the theory of BOHR, FRÖMAN and MOTTELSON<sup>36</sup> using the constants\*  $C_2=0.59$ ,  $C_4=0.005$  and  $C_6=0.0013$ . In Table 7 the experimental results are compared with the theoretical predictions.

Table 7. *Alpha group intensities in favoured alpha decay*

		$I=K_f$	$K_f+1$	$K_f+2$	$K_f+3$	$K_f+4$	$K_f+5$
Calculated	} relative intensities	100	13.7	2.2	0.017	0.0027	0.00023
		100	14.4	1.8	0.016	0.0033	0.00058

**4.3. The Level at 271 keV.** Apparently, this level must be assigned as  $3/2^-$   $1/2$  [530]. In agreement with previous authors no evidence was found for an alpha group associated with the intrinsic state  $1/2^-$   $1/2$  [530]. The experimental proof of this transition is made difficult by the fact that the most intense Am<sup>243</sup> group is close to the expected alpha particle energy.

The assignment  $M1$  made for the 210 keV transition (cf. Table 5) is in agreement with the results of other investigators.

As can be seen from Section 3, the 271 keV level is considerably populated by gamma transitions from higher excited states. This clarifies the large discrepancy between the 5278 keV alpha group intensity estimates based on fine structure measurements<sup>24, 25</sup> and on conversion electron studies<sup>23</sup>.

**4.4. The Rotational Band Based on the Intrinsic Level at 333 keV.** The results of the present investigation are consistent with the previous identification of a  $1/2^+$  [400] rotational band including energy levels at 333, 369, 373, 456 and 463 keV. The observed pattern of transitions from these levels is explained by the  $K$ -forbiddenness of dipole transitions and the strong hindrance of quadrupole transitions proceeding to the  $5/2^+$  [642] and  $5/2^-$  [523] rotational bands. Dipole decay to the level at 271 keV is also retarded, but is predicted to be competitive with the other transitions.

\* The  $C$ 's are the reciprocals of the hindrance factors found in neighbouring even-even nuclei.

<sup>36</sup> BOHR, A., P. O. FRÖMAN, and B. MOTTELSON: Dan. Mat. Fys. Medd. **29**, No.10 (1955).

**4.5. The Level at 360 keV.** BARANOV et al.<sup>25</sup> assigned to the 360 keV state the hole-excitation  $3/2^+$  [651] which should occur in this energy region. The alpha-gamma coincidence measurements of the present study indicate that the 360 keV level is predominantly de-excited by a ground state transition. This result is in favour of the above classification. According to the selection rules in the asymptotic quantum numbers the  $M 1$  transition to the configuration  $5/2^+$  [642] is unhindered. The weak 300 keV gamma ray may correspond to the strongly retarded  $E 1$  transition reaching the  $5/2^-$  [523] state.

**4.6. The Rotational Band Based on the Intrinsic Level at 440 keV.** The levels at 440, 489 and 550 keV fit well into a rotational band with the spin values  $3/2$ ,  $5/2$  and  $7/2$ . Two orbitals with spin  $3/2$  are predicted in this energy region, the hole-excitation  $3/2^-$  [532] and the particle-excitation  $3/2^-$  [521]. The results described in Section 3 point to a dominant population of the level at 271 keV. For the quantum numbers [532] the transitions to the states at 60 and 271 keV have to be classified as  $M 1 u$  and  $M 1 h$ , respectively. Obviously, the experimental data are in favour of the assignment [521]. The transitions in question then are  $M 1 h$  and  $M 1 u$ . Weak transitions to the ground state and to the level at 360 keV are expected due to strongly hindered  $E 1$  radiation. The classification given by BARANOV et al. is thus confirmed.

**4.7. The  $\beta$ -Vibrational Band.** The rotational band based on the level at 722 keV cannot be interpreted in terms of the Nilsson description. The small hindrance factors for the corresponding alpha groups indicate that these levels represent the  $\beta$ -vibrational band based on the intrinsic state at 60 keV. The present results confirm the data obtained by LEDE-  
RER<sup>26</sup> both regarding the de-excitation of the 722 keV level and the  $M 1$  character of the 660 keV radiation (cf. Table 5). As has already been pointed out by this author, the assignment of the 660 keV transition as  $M 1$  instead of  $E 2$  as well as the presence of relatively strong  $E 1$  radiation populating the ground state contradict definite predictions of the vibrational model. Apparently, the  $K$ -conversion coefficient for the new gamma ray at 690 keV indicates that this transition is  $M 1$ , too (cf. Table 5). We note, however, that the presumably lower intensity of the 650 keV transition proceeding to the 103 keV level (Fig. 6b) may point to a noticeable  $E 2$  admixture in these transitions. The ratio of reduced transition probabilities  $B(7/2 5/2 \rightarrow 7/2 5/2)/B(7/2 5/2 \rightarrow 5.2 5.2)$  is 1.86 for  $M 1$  radiation and 0.027 for  $E 2$  radiation.

**4.8. Level density.** Fig. 8 gives a comparison of the experimental intrinsic level excitation energies with the predictions of the Nilsson theory and the superfluid model. The level density calculated in terms of the superfluid model is approximately twice the level density yielded

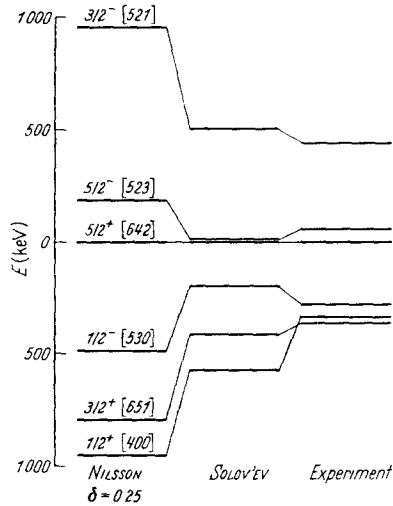


Fig. 8. Comparison of the experimental intrinsic level excitation energies with the predictions of the Nilsson theory and the superfluid model

by NILSSON's theory. Evidently, the experimental results justify the modifications introduced by SOLOV'EV. A similar result was recently reported by HORSCH<sup>37</sup> for the odd-N nucleus  $\text{U}^{235}$ .

<sup>37</sup> HORSCH, F.: Z. Physik **183**, 352 (1965).