KFK-322

KERNFORSCHUNGSZENTRUM

KARLSRUHE

März 1965

KFK 322

Gesellschaft für Kernforschung m.b.H.
Zentralbücherei
25. Juni 1965

Institut für Angewandte Kernphysik

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Zeitschrift für Physik 183, 352—374 (1965)

Aus dem Institut für Angewandte Kernphysik, Kernforschungszentrum Karlsruhe

Excited Energy Levels in U^{235}

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With 9 Figures in the Text

(Received November 25, 1964)

The transitions and levels in U^{235} populated by Pu^{239} alpha decay have been studied by alpha fine-structure investigations, alpha-gamma and gamma-gamma coincidence measurements. The spectra revealed the existence of at least 17 alpha groups and 24 gamma transitions which could be accounted for by 21 excited levels in U^{235}. The results are discussed within the framework of the unified model. The proposed level scheme exhibits a level density about twice as high as predicted by the Nilsson theory. This result is in fair agreement with the predictions of the superfluid model of SOLOV'EV.

1. Introduction

The nucleus U^{235} is in a region of strongly deformed nuclei. Consequently, its level scheme should exhibit pronounced rotational bands described by the Bohr-Mottelson nuclear model[1-3]. These rotational levels are based upon single-particle NILSSON states[4, 5]. More recently, SOLOV'EV[6, 7] has introduced some modifications in NILSSON's schemes by taking into account the effect of superfluidity. The calculated level density is about twice as large as that predicted by the Nilsson scheme. Experimental checking of the theoretical predictions is of great interest to nuclear physics. Moreover, knowledge of the U^{235} level scheme is a precondition for calculating the inelastic neutron scattering cross-section by means of the Hauser-Feshbach theory[8].

The levels in U^{235} are populated by the [9] 24413 y alpha decay of Pu^{239}. The alpha groups feeding levels higher than 53 keV are very

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[1] BOHR, A.: Dan. Mat.-Fys. Medd. **26**, No. 14 (1952).

[2] BOHR, A., and B.R. MOTTELSON: Dan. Mat.-Fys. Medd. **27**, No 16 (1953); — Phys. Rev. **89**, 316 (1953).

[3] BOHR, A., and B.R. MOTTELSON: Beta- and Gamma-Ray Spectroscopy (ed K. SIEGBAHN), Chapt. XVII. Amsterdam: North-Holland Publishing Company 1955

[4] NILSSON, S.G.: Dan. Mat.-Fys. Medd. **29**, No. 16 (1955).

[5] MOTTELSON, B.R., and S.G. NILSSON: Dan. Mat.-Fys. Skr. **1**, No. 8 (1959)

[6] SOLOV'EV, V. G.: Soviet Phys. JETP **13**, 456 (1961).

[7] VERESH, T., V.G. SOLOV'EV i T. SHIKLOSH: Bull. Acad Sci. U.S.S.R., Sér. phys. **26**, No 8, 1053 (1963).

[8] HAUSER, W., and H. FESHBACH: Phys. Rev. **87**, 366 (1952).

[9] LANDOLT-BORNSTEIN: Zahlenwerte und Funktionen aus Naturwissenschaft und Technik, Neue Ser., Gruppe 1 (ed. K.H. HELLWEGE). Berlin-Göttingen-Heidelberg· Springer 1961.

weak, the intensity being only 10^{-3} to 10^{-4}%. This causes significant experimental difficulties, especially for the alpha-gamma coincidence measurements described below. Such measurements have not been tried before.

The level structure of U^{235} has been the subject of several investigations [10-29]. Level schemes were proposed by Novikova et al.[13], Dzhelepov et al.[14], Baranov et al.[15], Newton[20], Shliagin[23], Tret'iakov et al.[24] and Murri and Cline[26]. On the basis of these works the existence of the [743] $\frac{7}{2}^-$ ground state[30-32] rotational band with members at 46 keV and 104 keV, of the [631] $\frac{1}{2}^+$ rotational band based upon an isomeric state[16-19, 33] at $\leqq 0.08$ keV with members at 13 keV, 53 keV, 83 keV and 152 keV, and of a $\frac{5}{2}^+$ level at 130 keV seems

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[10] Rosenblum, S., M. Valadares et B. Goldschmidt: Compt. rend. **230**, 638 (1950).

[11] Asaro, F., and I. Perlman: Phys Rev. **88**, 828 (1952).

[12] Gol'din, L.L., G.I. Novikova i E.F. Tret'iakov: Proc. Conf. Acad. Sci. USSR on Peaceful Uses of Atomic Energy, July 1955, Translation 174.

[13] Novikova, G.I., L.N. Kondrat'ev, Iu.P. Sobolev i L.L. Gol'din: Soviet Phys. JETP **5**, 832 (1957).

[14] Dzhelepov, B.S., R.B. Ivanov i V. G. Nedovesov: Soviet Phys. JETP **14**, 1227 (1962).

[15] Baranov, S.A., V. M. Kulakov i S.N. Belen'kii: Soviet Phys. JETP **16**, 801 (1963); — Nuclear Phys. **41**, 95 (1963).

[16] Michel, M.C., F. Asaro, and I. Perlman: Bull. Am. Phys. Soc. **2**, No. 2, 394 (1957).

[17] Freedman, M.S., F.T. Porter, F.Wagber, and P. Day: Phys. Rev. **108**, 836 (1957).

[18] Huizenga, J.R., C.L. Rao, and D.W. Engelkemeir: Phys. Rev. **107**, 319 (1957).

[19] Asaro, F., and I. Perlman: Phys. Rev. **107**, 318 (1957).

[20] Newton, J.O.: Nuclear Phys. **3**, 345 (1957).

[21] West, D., and J.K. Dawson: Proc. Phys. Soc. (London) A **64**, 586 (1951).

[22] Freedman, M.S., F.Wagner jr., and D.W. Engelkemeir: Phys. Rev. **88**, 1155 (1952).

[23] Shliagin, K.N.: Soviet Phys. JETP **3**, 663 (1956).

[24] Tret'iakov, E.F., G.I. Grishuk i L.L. Gol'din: Soviet Phys. JETP **7**, 560 (1958).

[25] Asaro, F., F.S. Stephens, and I. Perlman: Unpublished data (1957), cit. in ref. 23.

[26] Murri, E.L., and J.E. Cline: IDO 16695, 39 (1961).

[27] Asaro, F., and I. Perlman: Unpublished data, cit. in ref. 28 and 29.

[28] Perlman, I., and J.O. Rasmussen: Alpha Radioactivity, UCRL 3424 (1956).

[29] Perlman, I., u. J.O. Rasmussen: Handbuch der Physik, Bd. 42, S. 109. Berlin-Göttingen-Heidelberg: Springer 1957.

[30] Sluis, K.L. van der, and J.R. McNally: J. Opt. Soc. Am. **45**, 65 (1955).

[31] Hutchison jr., C.A., P.M. Llewellyn, E.Wong, and P. Dorain: Phys. Rev. **102**, 292 (1956).

[32] Blaise, J., S. Gerstenkorn et M. Louvegnies: J. phys. radium **18**, 318 (1957).

[33] Bohr, A., P.O. Froman, and B.R. Mottelson: Dan. Mat.-Fys. Medd. **29**, No. 10 (1955).

to be well established. Several studies show agreement in proposing levels at about 172 keV, 200 keV, 229 keV, 297 keV, 333 keV, 368 keV and 424 keV. Considerable discrepancies exist as to the existence or position of most gamma transitions in the decay scheme. The theoretical interpretation of several levels is thus mainly based on energy considerations. MURRI and CLINE observed 16 gamma transitions, only two of which, however, can be identified with transitions in the level scheme proposed by TRET'IAKOV et al. which contains 10 transitions. There is also disagreement concerning the spin assignment of the 172 keV level. TRET'IAKOV et al. found a spin value of $\frac{7}{2}$ and a parity equal to that of the ground state. MURRI and CLINE interpreted the same level as a rotational state to the 130 keV-$\frac{5}{2}^+$ intrinsic level they had introduced. Thus, they also obtained a spin value of $\frac{7}{2}$, but a parity opposite to that of the ground state. DZHELEPOV et al.[14] and BARANOV et al.[15] assumed this level to belong to the ground state rotational band and assigned to it $\frac{13}{2}^-$. BARANOV et al. found for the $(K=\frac{5}{2}^+)$ rotational band a level sequence with a rotational spacing factor of 4.4 keV which is markedly lower than the corresponding values of the ground state and $(K=\frac{1}{2}^+)$ rotational band (cf. Table 7). This implies an unusually large change of inertia. BARANOV et al. observed additional alpha groups feeding levels at 426 keV, 470 keV, 530 keV and possibly 93 keV. No interpretation of the levels at 224 keV, 333 keV, 424 keV, 426 keV, 470 keV and 530 keV has yet been attempted.

In order to clarify the contradictions mentioned above and to build up a more consistent level scheme it seemed necessary to reinvestigate both the alpha and gamma spectra. The rapidly advancing development of semiconductor detectors[34-37] furnished the decisive experimental material to work on.

2. Experimental Procedure

The U^{235} level structure was investigated by studying the fine structure of the alpha spectrum and by performing alpha-gamma coincidence and gamma-gamma coincidence measurements.

In the alpha-gamma coincidence equipment a 25 mm^2 semiconductor detector was used with about 100 μ depletion depth at 50 V bias. The total resolution including amplifier noise and energy loss in the source was about 25 keV. For gamma spectroscopy a $1\frac{1}{2}''\varnothing \times 1''$ NaI (Tl) crystal mounted on an EMI 6097 A photomultiplier was employed. The

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[34] Trans. IRE NS-8, No. 1 (1961) and NS-9, No. 3 (1962).

[35] NAS-NRC Publication 871, Nucl. Sci. Report No. 32 (1961).

[36] CZULIUS, W., H.D. ENGLER u. H. KUCKUCK: Ergebnisse der exakten Naturwissenschaften (ed. S. FLUGGE and F. TRENDELENBURG). Berlin-Göttingen-Heidelberg: Springer 1962.

[37] Nucleonics **20**, No. 5, 53 (1962).

energy resolution was 7.4% for the 662 keV gamma ray of Cs^{137}. Alpha fine structure measurements were performed by using an integral discriminator in the gamma pulse channel. Alpha-gamma coincidence measurements were carried out by setting a single channel pulse height discriminator on the various alpha groups. The main features of the electronic equipment are described elsewhere[38]. Because of the poor source strength in the alpha-gamma coincidence measurements a coincidence resolving time of 0.4 μs was sufficient. Data were recorded by a 256 channel pulse height analyser.

In the two-dimensional gamma-gamma coincidence arrangement two identical detectors were employed consisting of $1\frac{1}{2}'' \varnothing \times 2''$ NaI (Tl) crystals mounted on RCA 6810 A photomultipliers. The resolution was 8.1% and 8.4%, respectively, for the Cs^{137} gamma ray. A Compton shield[39] was used to avoid counter-to-counter scattering. For this arrangement a fast-slow coincidence circuit was chosen containing a 60 ns Bell-type fast coincidence circuit[40, 41]. Data were analysed by means of a 2 × 10 bit dual ADC with an integral linearity of 0.1% and a differential linearity of 1%. The digital data were processed and stored via one of the input channels of the CDC 160 A computer installed[42] at the Karlsruhe research reactor.

Sources were prepared by drying up a 1 n $HClO_4$ solution containing $2.1 \cdot 10^{-3}$ mol/l Pu^{239} onto a 0.2 mm fire-proof glass backing. These samples were heated in order to get a plutonium oxide layer as pure and uniform as possible. The active surface density of the alpha source was about 30 μg/cm² providing an activity of about 1 μC. The sample used for gamma-gamma measurements contained 1 mg of plutonium corresponding to an alpha activity of about 64 μC. The isotopic composition of the active material was $(96.8 \mp 0.2)\%$ Pu^{239}, $(3.2 \pm 0.2)\%$ Pu^{240}, $\leqq 0.2\%$ Pu^{241} and $\leqq 0.2\%$ Am^{241}. Shortly before this work was finished a source with 99.7% Pu^{239} became available*. Some control runs were performed with this source.

3. Experimental Results

3.1. Fine Structure of the Alpha Spectrum. In the total alpha spectrum (Fig. 1a) the most intense alpha groups of Pu^{239}, at 5147 keV and

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* From CBMN, Euratom, Geel, Belgium. The preparation is described in ref. 43

[38] Horsch, F.: Kernforschungszentrum Karlsruhe, Internal Report IAK No. 9/64 (1964).

[39] Bell, P. R.: Beta- und Gamma-Ray Spectroscopy (ed. K. Siegbahn), Chapt. V. Amsterdam: North-Holland Publishing Company 1955.

[40] Michaelis, W.: KFK 129 (1963).

[41] Bell, R. E., R. L. Graham, and H. E. Petch: Canad. J. Phys. **30**, 35 (1952)

[42] Kruger, G., and G. Dimmler: Int. Conf. on Nuclear Electronics, Paris. November 1963.

[43] Lauer, K. F., and V. Verdingh: Nuclear Instr. and Meth. **21**, 161 (1963).

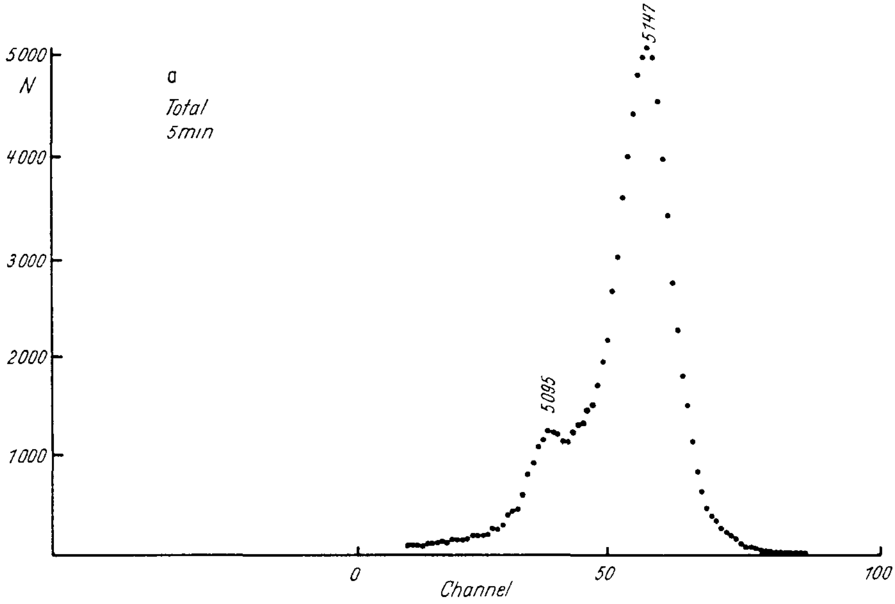

Fig. 1a–d. Alpha spectra

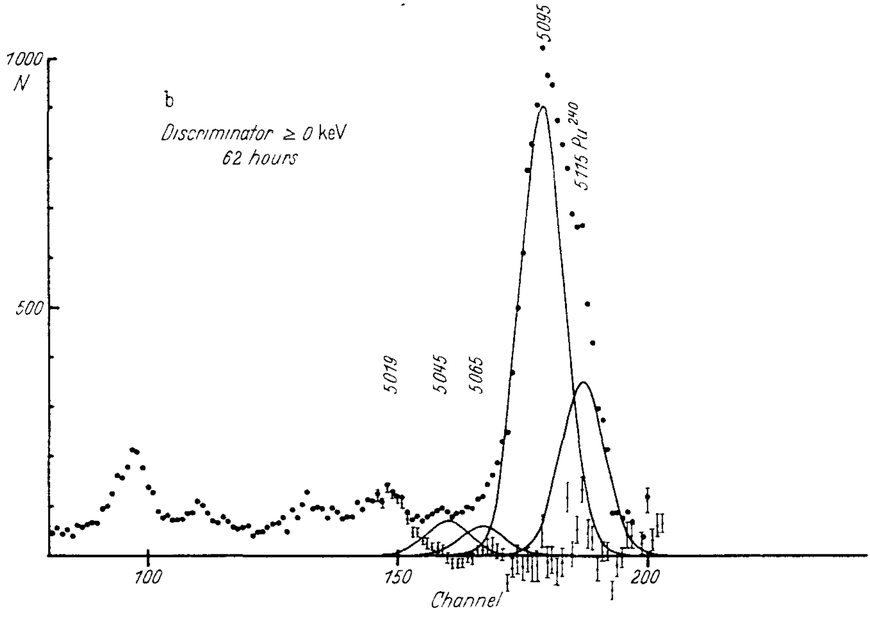

Fig. 1b

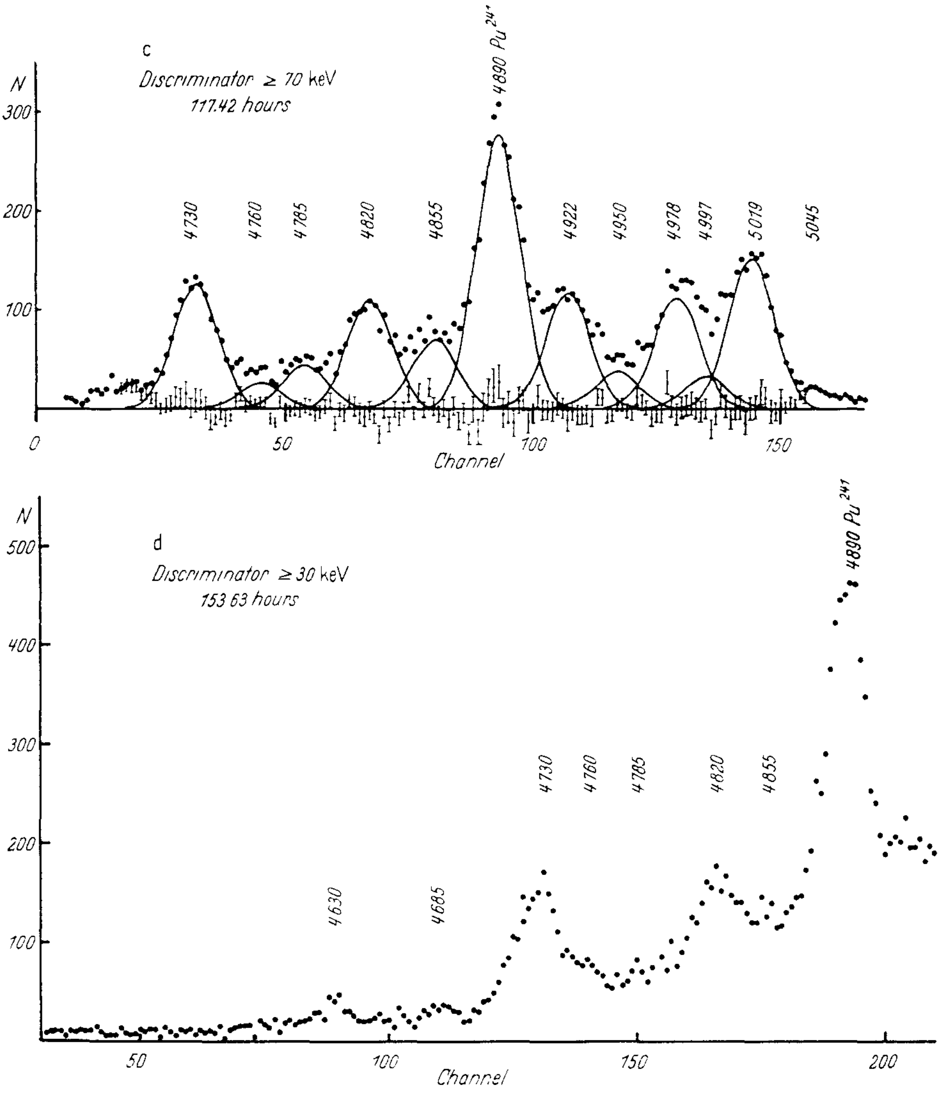

Fig. 1c u. d

5134 keV, and of Pu^{240}, at 5159 keV and 5115 keV[44], remain unresolved and produce a broad peak at 5147 keV. The low-energy tails of this peak and of the 5095 keV line mask all weaker alpha groups. The alpha-gamma coincidence method, however, permitted only such alpha groups to be detected which feed excited levels in the daughter nucleus.

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[44] Strominger, D., J. M. Hollander, and G. T. Seaborg: Rev. Mod. Phys. **30**, No. 2 (1958).

Consequently, the alpha spectrum could be unfolded step by step by rising the gamma discriminator level (Fig. 1 b – d). Employing this method even alpha groups with an intensity of about $10^{-4}\%$ were clearly observed. All alpha spectra are corrected for random coincidences.

3.2. Alpha-Gamma Coincidence Measurements. The results of the alpha-gamma measurements are displayed in Figs. 2 – 4. The 58 keV

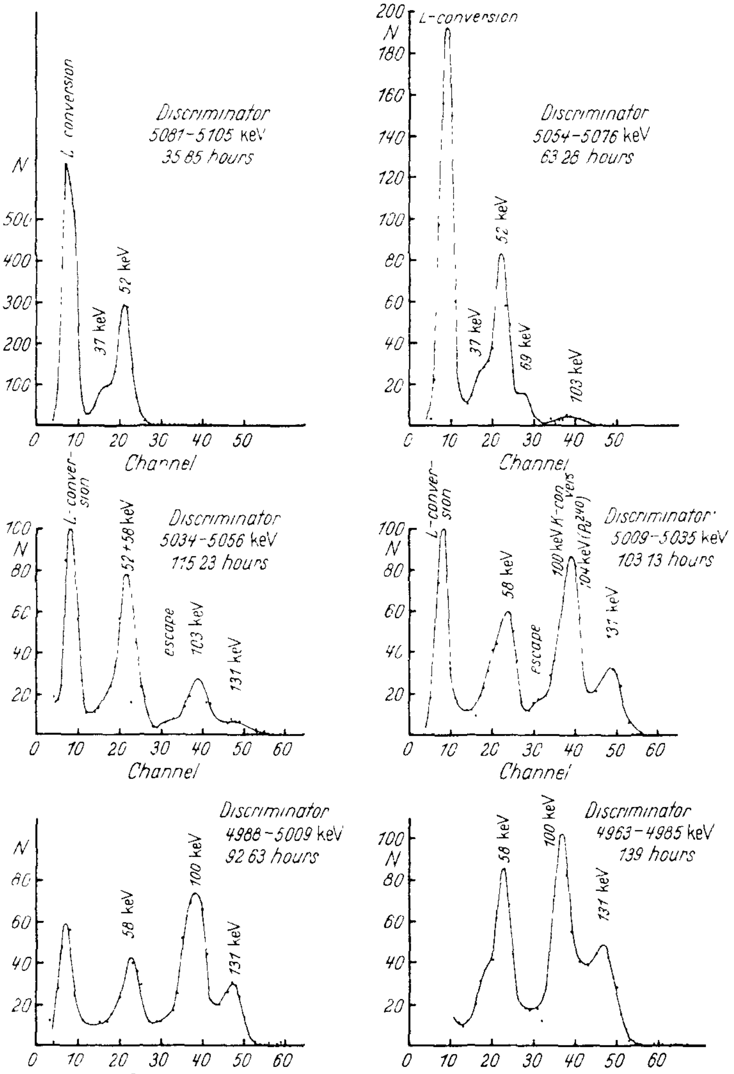

Fig. 2. Alpha-gamma coincidence spectra to the alpha energy range from 4963 to 5105 keV

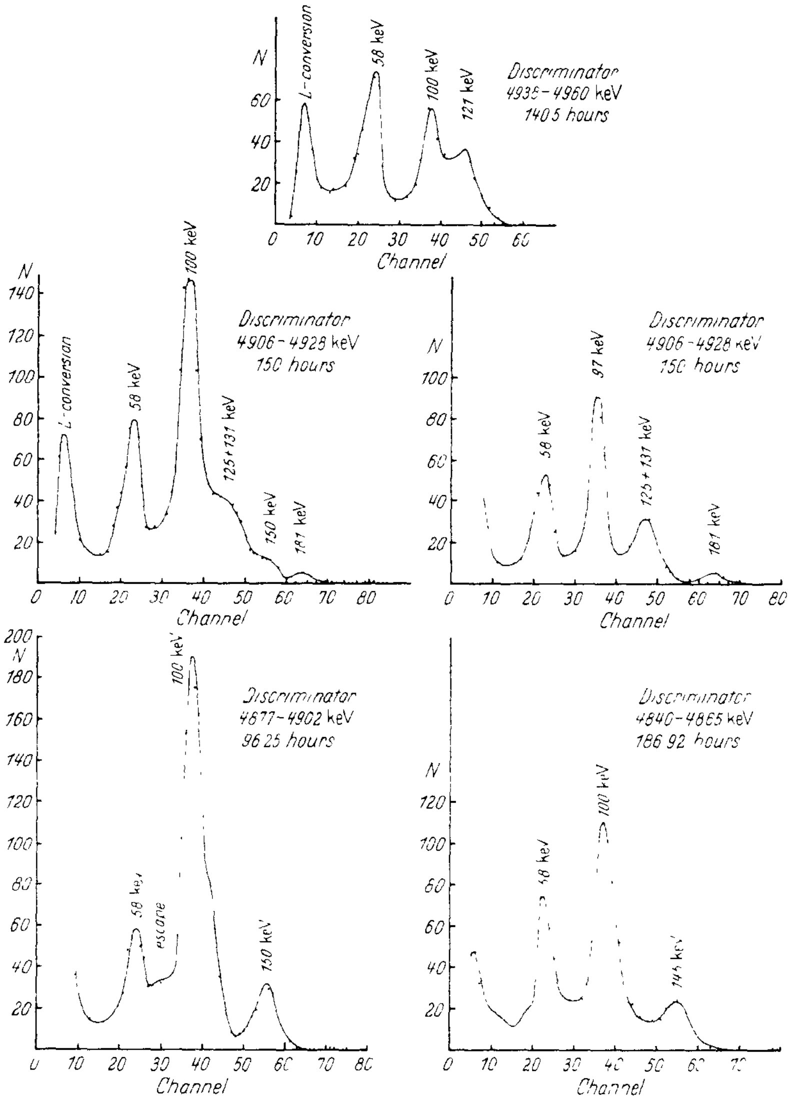

Fig. 3. Alpha-gamma coincidence spectra to the alpha energy range from 4840 to 4960 keV (the figure shows two coincidence spectra to the same alpha energy range from 4906 to 4928 keV, the second being corrected for contributions from neighbouring alpha groups)

line contains counts resulting from coincidences to the low-energy tails of the Pu^{239}-5095 keV and Am^{241}-5477 keV alpha groups[44] and from random coincidences. In the gamma spectrum coincident with the 4997 keV line, for instance, the various contributions were estimated

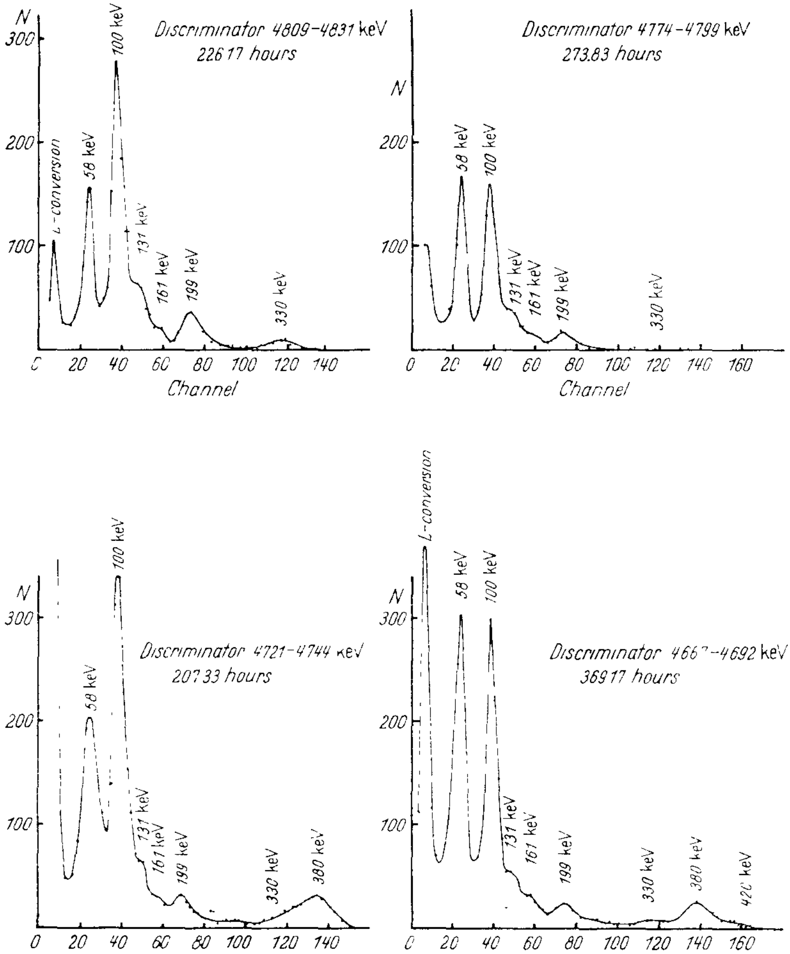

Fig. 4. Alpha-gamma coincidence spectra to the alpha energy range from 4667 to 4831 keV

to be 30:30:40. In the gamma spectrum coincident with the alpha energy range of 4877 keV – 4902 keV the 100 keV and 150 keV lines are due [22] to Pu^{241}. The results are summarized in Table 1. The coincidences given in parentheses possibly exist but cannot be definitely established from alpha-gamma spectra.

3.3. Gamma-Gamma Coincidence Measurements. The total gamma spectrum taken with a $1\frac{1}{2}''\varnothing \times 2''$-NaI(Tl) crystal is given in Fig. 5. Some typical coincidence spectra are shown in Figs. 6 and 7. They

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

Alpha energy range (keV)	Coincident gamma ray energies (keV) 37	52	58	69	97	K 100 103	121	125 127 130 131	145 150	161	181	199	230	330	380	420
5081–5105	+	+														
5054–5076	+	+		+		+										
5034–5056		+	+			+		+								
5009–5035			+			+		+								
4988–5009			+			+		+								
4963–4985			+			+		+								
4938–4960			+			+	+									
4906–4928			+		+	(+)		+			+		(+)			
4877–4902			+			+			+							
4840–4865			+			+		(+)	+							
4809–4831			+			+		+		+		+		+		
4774–4799			+			+		+		+		+		+		
4721–4744			+			+		+		+		+		+	+	
4667–4692			+			+		+		+		+		+	+	+

were analysed by the method proposed by PREUSS and ESCARFAIL[45]. The well-known broadening of a K *X*-ray in the analysis caused two lines at 100 keV and 114 keV. The "true" shape of the K X-rays in the spectra is thus represented by the sum of the K portion of the 100 keV peak and the 114 keV peak. Because of the long run time the gamma-gamma spectra could not be corrected for random coincidences. In all spectra the 58 keV line consists almost completely of random coincidences from the intense Am^{241}-$E1$ radiation[44] thus allowing an estimate of the contribution of random coincidences to other gamma energies from the total gamma spectrum. Therefore, the gamma-gamma spectra were handled carefully, and the gamma ray intensities were deduced from the alpha-gamma coincidence spectra, except for the values of the 165 keV, 189 keV, 245 keV, 260 keV and 294 keV gamma rays which may be less accurate. After a 52 h control run with a pure Am^{241} sample* the contribution from true coincidences of Am^{241} proved to be negligible. The results are summarized in Table 2.

3.4. Gamma Ray Energies and Intensities. Errors. The maximum estimated limits of accuracy for the gamma ray energies listed in Table 3 are ±5%.

For evaluating the gamma ray intensities from the alpha-gamma coincidence measurements the spectra had first to be corrected for con-

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* In order to prevent counting rate effects in the photomultipliers the Am^{241} gamma source strength was chosen equal to that of the Pu^{239} source.

[45] PREUSS, L. G., and J. P. ESCARFAIL: Nuclear Instr. and Meth. 9, 212 (1960).

tributions from neighbouring alpha groups. Especially the interference from the high-energy alpha groups was not negligible due to the low-energy tail characteristic for semiconductor detectors. All values given

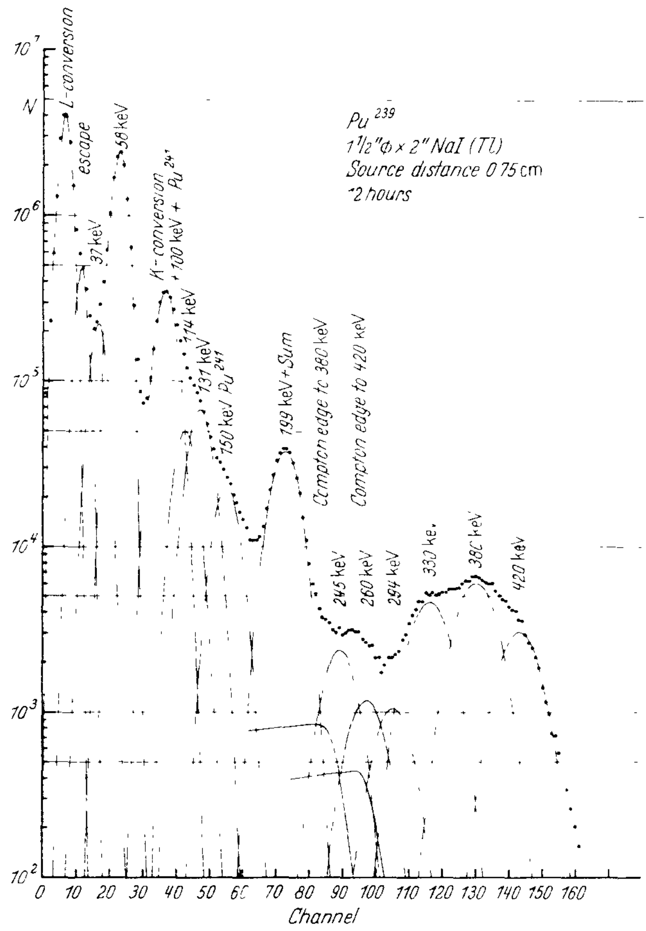

Fig. 5. Total gamma spectrum

in Table 3 are corrected for absorption*, peak-to-total ratio[46] and total efficiency[47]. The errors assigned to the intensities of unconverted photons include statistical errors, estimates of errors introduced in

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[*] 5/9/62 J. G. B., Harshaw Chemie GmbH., Frankfurt/Main.

[46] Rathburn, E. R., and C. E. Crouthamel: Applied Gamma-Ray Spectrometry (ed. C. E. Crouthamel). Oxford: Pergamon Press 1960.

[47] Mott, W. E., u. R. B. Sutton: Handbuch der Physik, Bd. 45, S. 86. Berlin-Göttingen-Heidelberg: Springer 1958.

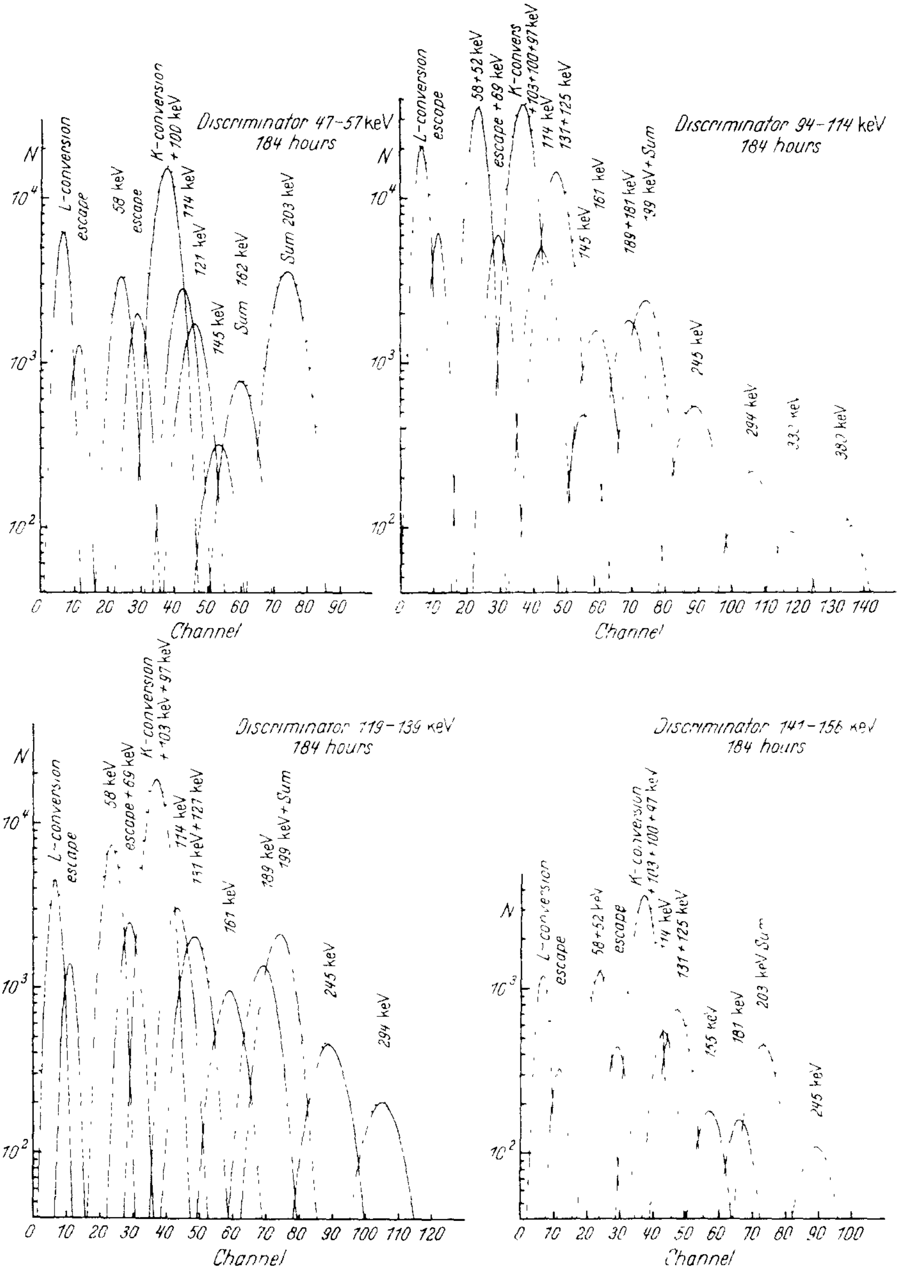

Fig. 6. Gamma-gamma coincidence spectra to the energy range from 47 to 156 keV

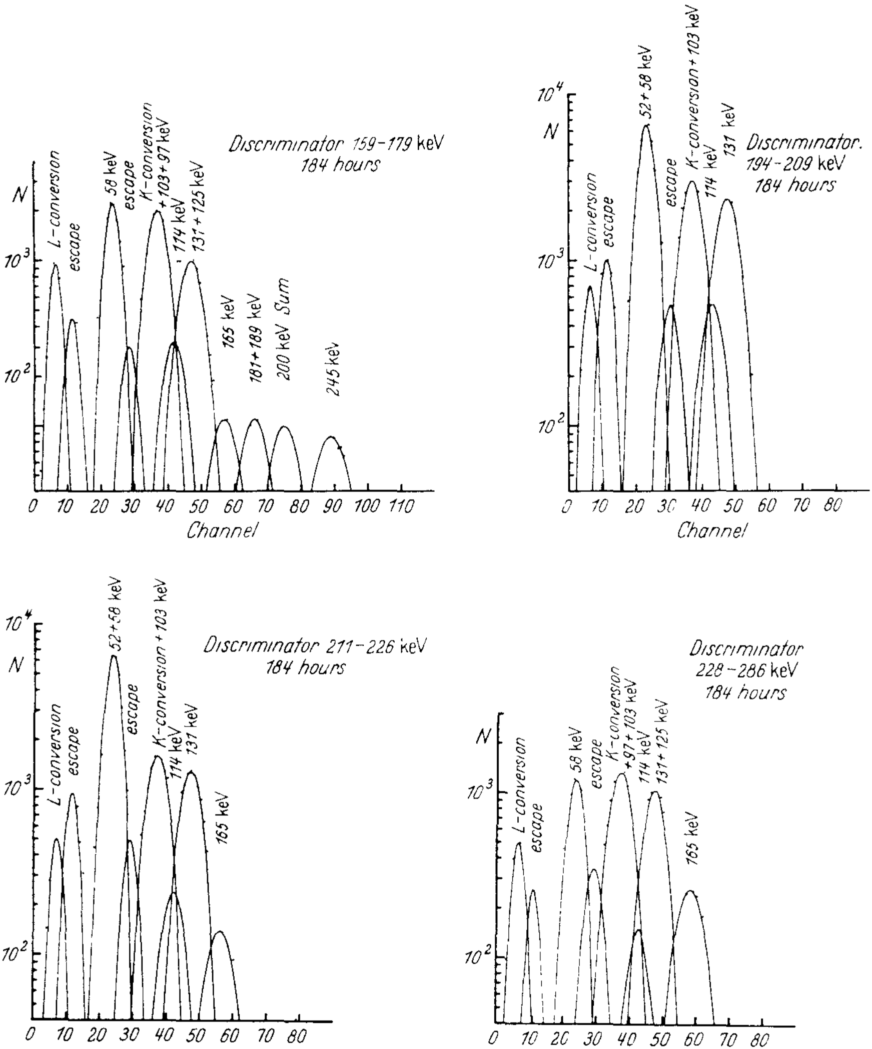

Fig. 7. Gamma-gamma coincidence spectra to the energy range from 159 to 286 keV

treating incompletely resolved lines and estimates of errors introduced in determining the contribution from the tails of nearby alpha lines. The uncertainties in the corrections for absorption, peak-to-total ratio and for total efficiency were negligible.

The gamma coincidence spectra of the 4730 keV and the 4685 keV alpha groups were measured with a different geometry. In order to reduce the run time the collimator in front of the semiconductor detector was enlarged from 3 mm ∅ to 4 mm ∅. In referring the gamma ray intensities to the values obtained with the 3 mm ∅ collimator, possible inhomogenities in the source layer were not considered. This effect may have introduced some further error into the intensities of the 131 keV, 161 keV, 199 keV, 330 keV and 380 keV lines.

Table 2. *Summary of the gamma-gamma coincidence measurements*

Gamma energy range	Coincident gamma ray energies (keV)										
(keV)	52 58	97 K 100 103	121 125 127 131	145	161 165	181 189	199	230 245	294	330	380
47– 57	+	+	+	+	+		+				
62– 77	+	+	+	+	+		+				
79– 91	+	+	+		+	+	+				
94–114	+	+	+	+	+	+	+	+	+	+	+
119–139	+	+	+		+	+	+	+	+		
141–156	+	+	+		+	+	+	+			
159–179	+	+	+		+	+	+	+			
194–209	+	+	+								
211–226	+	+	+		+						
228–286	+	+	+		+						
288–321	+	+	+								
323–356		+	+								
356–393		+									
395–428	+	+									
431–468		+									

3.5. Alpha Energies and Intensities. Errors. The limit of accuracy of the alpha energies at 5095 keV, 5065 keV, 5045 keV, 5019 keV, 4978 keV, 4922 keV, 4890 keV, 4820 keV and 4730 keV listed in Table 4 is estimated to be ±4 keV, that of the alpha energies at 4997 keV, 4950 keV, 4855 keV, 4785 keV, 4760 keV, 4685 keV and 4630 keV is estimated to be ±8 keV. The same errors apply to the level energies in Table 4 which were calculated from alpha energies. The 5147 keV group* of Pu^{239} and the 4816 keV group[9] of U^{233} were used as standards.

Employing experimental or theoretical K- and L-conversion coefficients[48, 49] the alpha intensities were calculated from the gamma ray intensities and thus contain the errors mentioned in subsection 3.4. The K-conversion coefficients by Rose et al. and the L-conversion coefficients

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* Baranov gives a value of 5157 keV.

[48] Rose, M.E., G.H. Goertzel, and C.L. Perry: K-shell Internal Conversion Coefficient Revised Tables, ORNL 1023.

[49] Sliv, L.A., and I.M. Band: Coefficients of Internal Conversion of Gamma-Radiation, Part 2, L-shell, Translation in NPTR 217 (1958).

by SLIV and BAND have an uncertainty of ±2% which is estimated to be increased by interpolating up to ±5%. However, this uncertainty only introduced a remarkable contribution into the error for the 5095 keV alpha group. The 5134 keV and 5147 keV alpha groups could not be resolved. Consequently, Table 4 gives only the values found by BARANOV et al. In agreement with these authors no evidence for an alpha transition to the 46 keV level, which had been observed in Coulomb excitation measurements, was found in any of the alpha and alpha-gamma coincidence spectra.

Table 3. *Gamma ray intensities*

E_γ (keV)	Relative intensity [a]	Absolute intensity (%) [b]
	of unconverted photons	
37	65 ± 11	$(2.3\pm0.4)\cdot10^{-2}$
52	100 ± 5	$(3.6\pm0.3)\cdot10^{-2}$
69	2.2 ± 0.7	$(8.0\pm2.4)\cdot10^{-4}$
97	$\leqq 2.7$	$\leqq 1.0\cdot10^{-3}$
100	2.9 ± 1.6	$(1.1\pm0.6)\cdot10^{-3}$
100 Pu^{241}	25 ± 3	
103	1.5 ± 0.7	$(5.3\pm2.4)\cdot10^{-4}$
K	34 ± 3	$(1.2\pm0.1)\cdot10^{-2}$
121	2.1 ± 0.7	$(7.5\pm2.5)\cdot10^{-4}$
125	16 ± 1.1	$(5.7\pm0.5)\cdot10^{-3}$
127		
130		
131		
145	0.7 ± 0.4	$(2.3\pm1.5)\cdot10^{-4}$
150 Pu^{241}	4.0 ± 0.8	
161 + Sum	1.4 ± 0.2	$(5.0\pm0.8)\cdot10^{-4}$
165	0.5 ± 0.3 [c]	$(1.8\pm1.0)\cdot10^{-4}$
181	0.3 ± 0.15	$(1.1\pm0.5)\cdot10^{-4}$
189	0.27 ± 0.15 [c]	$(1.0\pm0.5)\cdot10^{-4}$
199 + Sum	3.6 ± 0.5	$(1.3\pm0.2)\cdot10^{-3}$
230	$\leqq 0.035$	$\leqq 1.2\cdot10^{-5}$
245	0.7 ± 0.15 [c]	$(2.6\pm0.6)\cdot10^{-4}$
260	0.42 ± 0.19 [c]	$(1.5\pm0.7)\ 10^{-4}$
294	0.46 ± 0.08 [c]	$(1.6\pm0.3)\cdot10^{-4}$
330	2.6 ± 0.5	$(9.1\pm1.8)\cdot10^{-4}$
380	3.7 ± 0.7	$(1.3\pm0.3).10^{-3}$
420	2.1 ± 0.4	$(7.5\pm1.5)\cdot10^{-4}$

a) Referred to an arbitrary value of 100 for the 52 keV line.

b) Referred to an absolute intensity of the 5095 keV alpha group of 11.5%.

c) Deduced from the total gamma spectrum and from the gamma-gamma coincidence spectra; not visible in the alpha-gamma coincidence spectra.

3.6. Multipolarities. *K*-Conversion Coefficients. An attempt was made at determining the mixing ratio of the 37 keV transition from the $(\frac{1}{2}, \frac{5}{2}^+)$ level to the $(\frac{1}{2}, \frac{3}{2}^+)$ level using the relative intensities of the 37 keV and the 52 keV lines and the theoretical value of the reduced transition probabilities (cf. Table 5). According to TRET'IAKOV et al. the 52 keV line is pure $E2$ radiation. For the theoretical L-conversion coefficients the values given by SLIV and BAND were used. The result of 96% $M1$ + 4% $E2$ considerably differs from the mixing ratio 87% $M1$ + 13% $E2$ reported by TRET'IAKOV et al.

The K-conversion coefficient of the 131 keV transition was obtained from the gamma coincidence spectrum to the 5019 keV alpha group. Its value 1,5 is 20% smaller than that measured by MURRI and CLINE.

Table 4. *Fine structure of Pu^{239} alpha spectrum*

Excited level in U^{235} (keV)	Alpha energy (keV)	Relative	Absolute (%)	Hindrance factor F	Values of BARANOV (%)
		intensity			
0	5147				73.3
0.08					
13	5134				15.1
46	—				$<3 \cdot 10^{-2}$
53	5095	84350 ± 4350	11.5 [a]		11.5
83	5065	310 ± 100	$(4.3 \pm 1.4) \cdot 10^{-2}$	1220 ± 400	$3.2 \cdot 10^{-2}$
104	5045	$\geqq 24$	$\geqq 3.3 \cdot 10^{-3}$	$\leqq 11500$	$2.1 \cdot 10^{-2}$
130	5019	28 ± 5	$(3.8 \pm 0.7) \cdot 10^{-3}$	8200 ± 1450	$5 \cdot 10^{-3}$
152	4997	82 ± 46	$(1.1 \pm 0.6) \cdot 10^{-2}$	2200 ± 1250	$8 \cdot 10^{-3}$
172 $\frac{7}{2}^{+}$ [b] $\frac{13}{2}^{-}$	4978	30 ± 3	$(4.1 \pm 0.5) \cdot 10^{-3}$	3950 ± 400	$5 \cdot 10^{-3}$
200	4950	32 ± 8	$(4.4 \pm 1.1) \cdot 10^{-3}$	2450 ± 600	$3 \cdot 10^{-3}$
229	4922	21 ± 4	$(2.9 \pm 0.6) \cdot 10^{-3}$	2200 ± 420	$3 \cdot 10^{-3}$
Pu^{241}	4890	75 ± 8			
297 $\frac{13}{2}^{+}$ $(\frac{11}{2}^{+})$	4855	5 ± 3	$(7 \pm 4) \cdot 10^{-4}$	3800 ± 2300	$7 \cdot 10^{-4}$
333	4820	15.5 ± 2.5	$(2.1 \pm 0.4) \cdot 10^{-3}$	860 ± 140	$1.5 \cdot 10^{-3}$
368	4785	6.0 ± 1.5	$(8 \pm 2) \cdot 10^{-4}$	1200 ± 300	$6 \cdot 10^{-4}$
394	4760	$\leqq 7$	$\leqq 1.0 \cdot 10^{-3}$	$\geqq 590$	—
424 $\frac{9}{2}^{+}$ (?)	4730	22 ± 3	$(3.0 \pm 0.4) \cdot 10^{-3}$	110 ± 15	$2.6 \cdot 10^{-3}$
470	4685	$\leqq 4.5$	$\leqq 6 \cdot 10^{-4}$	$\geqq 300$	$4 \cdot 10^{-4}$
526	4630	$\leqq 5.5$	$\leqq 8 \cdot 10^{-4}$	$\geqq 85$	$2 \cdot 10^{-4}$

a) Reference value for all other absolute intensities.

b) The quantum numbers in parenthesis shall indicate the existence of ever two levels at 172, 297 and 424 keV.

Table 5. *K-conversion coefficients. Multipolarities*

Gamma ray energy (keV)	K-conversion coefficient	Multipolarity
37		$96\%\ M1 + 4\%\ E2$ [a]
131	1.5 ± 0.35	$(96.6 \pm 0.9)\%\ E1$ $+ (3.4 \mp 0.9)\%\ M2$
189 380	0.43 ± 0.40	b

a) From the intensity ratio to the 52 keV $E2$ transition (cf. text).

b) On the basis of the intensities given in Table 3 the theoretical K-conversion coefficient for the multipolarities 189 keV—$M1$ and 380 keV—$E1$ has a value of 0.045 ± 0.025.

Table 6. *Experimental and theoretical reduced transition probabilities*

Energy of initial state (keV)	Reduced transition probability	Theory	Experiment
229	$B(E1; \frac{9}{2}^{+} \to \frac{11}{2}^{-})$ $B(E1; \frac{9}{2}^{+} \to \frac{9}{2}^{-})$	2.03	$\leqq 170$ [a]
229	$B(E1; \frac{9}{2}^{+} \to \frac{9}{2}^{-})$ $B(E1; \frac{9}{2}^{+} \to \frac{7}{2}^{-})$	14.56	$\geqq 9.1$
333	$B(M1; \frac{5}{2}^{+} \to \frac{7}{2}^{+})$ $B(M1; \frac{5}{2}^{+} \to \frac{5}{2}^{+})$	0.40	1.08 [b]
424	$B(M1; \frac{9}{2}^{+} \to \frac{9}{2}^{+})$ $B(M1; \frac{9}{2}^{+} \to \frac{7}{2}^{+})$	0.82	2.8 ± 2.1

a) Deduced from the intensity of the (125+131) keV peak.

b) Falsified by sum contributions both in the 161 keV and the 199 keV line.

From this result the mixing ratio of the 131 keV radiation was calculated employing the K-conversion coefficients tabulated by Rose et al.

The total conversion coefficient of the 189 keV and 380 keV lines was obtained from the gamma spectrum concident with the 4730 keV alpha group, from the 4820 keV coincidence spectrum using the intensity ratio of the K X-rays to the (161 keV+sum), (199 keV+sum) and 330 keV radiation.

3.7. Reduced Transition Probabilities. In Table 6 some experimental reduced transition probabilities are compared with the theoretical values (cf. sect. 4).

4. Discussion

4.1. Level Scheme. All transitions identified in these experiments are explained by the level scheme shown in Fig. 8. The investigation both of alpha-gamma and gamma-gamma coincidence relationships provides convincing evidence of the correctness of this decay scheme. The transitions in Fig. 8 are divided into transitions within rotational bands and into transitions between levels belonging to different rotational bands. The dashed levels and transitions probably exist, but could not be definitely established because of the relatively poor intensity of the corresponding radiation.

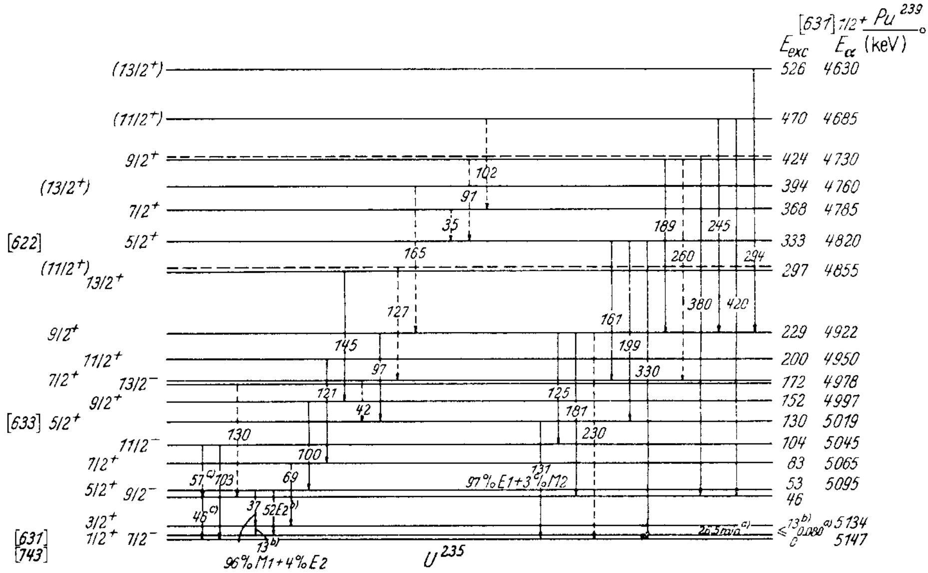

Fig. 8. Level scheme of U^{235} based on the results of the present investigations. Theoretical Interpretation [$Nn_z\Lambda$] $I\pi$, a) acc. to M. C. Michel et al [16], b) acc. to E. F. Tretjakov et al [24], c) acc. to J. O. Newton [20]

The level sequence up to 152 keV agrees with that proposed by Baranov et al. Within this range the levels can be assigned to a $(K=\frac{7}{2}^{-})$ ground state rotational band, a $(K=\frac{1}{2}^{+})$ rotational band based on the isomeric 0.08 keV level and a $(\frac{5}{2}, \frac{5}{2}^{+})$ intrinsic level at 130 keV.

The experimental results are theoretically interpreted in the following subsections.

4.2. The Ground State Rotational Band. Of the $(K=\frac{7}{2}^{-})$ ground state rotational band the first two members at 46 keV and 104 keV seem to be well established. According to the predictions of the simple rotational formula

$$E_{rot}=\frac{\hbar^2}{2\Theta}\{I(I+1)-K(K+1)\} \qquad I=K,\ K+1,\ K+2,\dots.$$

Dzhelepov et al. and Baranov et al. interpreted the level at 172 keV, which was first reported by Asaro and Perlman[27], as the third member of the $(K=\frac{7}{2}^{-})$ band. In the 4978 keV alpha-gamma coincidence spectrum the intensity ratio of the K X-rays and the 131 keV radiation (cf. subsect. 4.4) seems to be shifted compared with the 5019 keV coincidence spectrum. This may be explained by the presence of another 130 keV transition. If this assumption is correct, the interpretation given by Dzhelepov et al. and Baranov et al. is confirmed. The 46 keV transition from the $(\frac{7}{2}, \frac{9}{2}^{-})$ level to the ground state is highly L-converted and consequently not visible in the gamma ray spectrum. However, the intensity of the 4978 keV alpha group can hardly be explained only by the $(\frac{7}{2}, \frac{13}{2}^{-})$ level because the large spin difference to the Pu^{239} ground state and the parity change require an alpha particle angular momentum of $L\geqq 7$. This problem might be clarified by the results discussed below which suggest a second level at 172 keV with the quantum numbers $(\frac{5}{2}, \frac{7}{2}^{+})$.

4.3. The Rotational Band Based on the Intrinsic Level at $\leqq$0.08 keV. The rotational levels with spins $\frac{3}{2}^{+}$, $\frac{5}{2}^{+}$, $\frac{7}{2}^{+}$ and $\frac{9}{2}^{+}$ are well established. From alpha fine-structure investigations Dzhelepov et al. and Baranov et al. proposed the higher members $\frac{11}{2}^{+}$ and $\frac{13}{2}^{+}$ at 200 keV and 297 keV, respectively. The present experiments revealed the hitherto unknown $E2$ transitions proceeding from these levels to the $(\frac{1}{2}, \frac{7}{2}^{+})$ and $(\frac{1}{2}, \frac{9}{2}^{+})$ members. The assignments $(\frac{1}{2}, \frac{11}{2}^{+})$ and $(\frac{1}{2}, \frac{13}{2}^{+})$ are thus confirmed. The interpretation is supported by the fact that no transitions to the lower members of this band and to the $(K=\frac{7}{2}^{-})$ band are observed. Dipole and quadrupole transitions to the ground state rotational band are forbidden because of the large K difference.

4.4. The Rotational Band Based on the Intrinsic Level at 130 keV. In gamma-gamma coincidence measurements Murri and Cline found the first indications* of a level at 130 keV. From analogy conclusions to the Pu^{239} level scheme and as a consequence of the 130 keV K-conversion coefficient obtained from their gamma coincidence spectrum to the 207 keV gamma ray they assigned this state [622] $\frac{5}{2}^+$. The conversion coefficient given in Table 5 supports the interpretation of these authors concerning spin and parity. However, the classification [622] is uncertain. According to the Nilsson theory as well as to the superfluid model this orbital is expected to occur at a higher energy than the [633] $\frac{5}{2}^+$ hole state. Indeed, the present experiments indicate the existence of an additional $\frac{5}{2}^+$ single particle state at 333 keV. The 333 keV level must be interpreted as the state [622] on the basis of the intensity ratios of the 161 keV, 199 keV and 330 keV gamma rays. Only a strong hindrance of the 330 keV-$E1$ transition according to the assymptotic quantum number selection rules can explain its relative intensity to the 161 keV-$M1$ and the 199 keV-$M1$ radiations. The classification [622] causes a stronger hindrance[5] of the 330 keV-$E1$ transition than the assignment [633]. Consequently, there is some evidence of the 130 keV level corresponding to the [633] $\frac{5}{2}^+$ hole state. This assignment can also account for the measurable $M2$ admixture of the 131 keV-$E1$ radiation, the $E1$ radiation being hindered, the $M2$ radiation, however, being unhindered.

From conversion electron investigations TRET'IAKOV et al. reported two $E1$ transitions from the 172 keV level to the $(\frac{1}{2}, \frac{7}{2}^+)$ and $(\frac{1}{2}, \frac{5}{2}^+)$ levels. As a consequence, these authors ascribed to this level a spin value of $\frac{7}{2}$ and a parity opposite to that of the $(K=\frac{1}{2}^+)$ band. Thorough investigation of the spectra discussed in section 3 revealed no gamma rays which might correspond to the transitions observed by TRET'IAKOV et al. The spectra point to a strong coincidence relationship of the 4978 keV alpha group and the 131 keV line which also appears in the 5019 keV coincidence spectrum. Alpha intensity considerations provide strong evidence that the cited gamma intensity cannot be explained just by a 130 keV-$E2$ transition within the ground state rotational band. The experimental result can be interpreted by an additional $(\frac{5}{2}, \frac{7}{2}^+)$ level at 172 keV. According to the predictions of the rotational formula the $(\frac{5}{2}, \frac{9}{2}^+)$ level is expected to occur at about 226 keV. Within experimental errors this prediction agrees with the level at 229 keV revealed by the alpha spectrum. Indeed, the reduced transition probabilities of the 125 keV, 181 keV and 230 keV gamma rays are in favour of a spin value $\frac{9}{2}$ (cf. Table 6). The above arguments support the conclusion that the first and the second [633] rotational levels lie at 172 keV and 229 keV. This result is in contrast to the $(K=\frac{5}{2}^+)$ rotational band proposed by BARANOV et al. The level sequence 130 keV, 160 keV, 204 keV and

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* A strong 130 keV—207 keV coincidence relationship.

248 keV given by these authors yields a rotational spacing factor of about 4.4 keV. This value considerably deviates from the spacing factors for the other U^{235} rotational bands (cf. Table 7). The level sequence obtained in this work gives a rotational spacing factor of 6.2 keV. The values[5] 6.08 keV, 6.0 keV and 5.83 keV for the same single particle configuration [633] $\frac{5}{2}^+$ in the nuclei Th^{229}, Th^{231} and U^{233}, respectively, are in favour of our result. In the present experiments no indications were found for levels at 160 keV, 204 keV and 248 keV. The third and the fourth members of the [633] $\frac{5}{2}^+$ band with spins $\frac{11}{2}^+$ and $\frac{13}{2}^+$ are predicted to occur at about 297 keV and 380 keV. In fact, BARANOV'S alpha spectrum exhibits a double peak at an alpha energy which corresponds to the 297 keV level suggesting a second level at this energy. The weak 131 keV – 127 keV coincidence relationship revealed in the spectra of section 3 might arise from the expected $E2$ transition to the $(\frac{5}{2}, \frac{7}{2}^+)$ level. The alpha spectrum (cf. Fig. 1) furthermore shows a weak line at about 4760 keV corresponding to an U^{235} level at 394 keV unknown up till now. Considering the uncertainties discussed in section 3 this energy nearly agrees with the predicted energy for a $(\frac{5}{2}, \frac{13}{2}^+)$ level. Hence, the weak 165 keV line found in coincidence with the 181 keV and 230 keV transitions would correspond to an $E2$ transition to the $(\frac{5}{2}, \frac{9}{2}^+)$ level.

Table 7. *Rotational spacing factors in U^{235}*

Intrinsic level in U^{235}	Rotational spacing factor $\frac{\hbar^2}{2\Theta}$ (keV)	Decoupling parameter (keV)
[743] $\frac{7}{2}^-$	5.2	–
[631] $\frac{1}{2}^+$	5.98	−0.235
[633] $\frac{5}{2}^+$	6.2	–
[622] $\frac{5}{2}^+$	5.2	–

4.5. The Rotational Band Based on the Intrinsic Level at 333 keV. The levels at 333 keV, 368 keV, 424 keV, 470 keV and 526 keV are still left for interpretation. The fair agreement of the level energies obtained in the present work with the values found by BARANOV et al. support their accuracy. These levels do not fit into the first three rotational bands discussed above, apart from the fact that the relatively high intensity of the corresponding alpha groups is incompatible with spin values $\geqq \frac{15}{2}$. In the 4820 keV alpha-gamma coincidence spectrum three new gamma lines appear which are ascribed to transitions proceeding to the levels $(\frac{5}{2}, \frac{5}{2}^+)$, $(\frac{5}{2}, \frac{7}{2}^+)$ and to the ground state or isomeric state at $\leqq 0.08$ keV. This pattern suggests the 333 keV level to be a new intrinsic state. The interpretation is supported by the similarity of the coincidence spectra to the alpha groups at 4820 keV, 4785 keV, 4730 keV and 4685 keV (a coincidence spectrum to the 4630 keV alpha group has not been taken because of the poor intensity). All spectra indicate gamma rays at 161 keV, 199 keV and 330 keV. This fact suggests the conclusion that the levels at 368 keV, 424 keV and 470 keV are rotational levels based on the 333 keV intrinsic configuration. Most probably, $M1$ and $E2$ transitions within the band cause the higher levels to be de-excited via the 333 keV state.

Within the framework of the Nilsson theory the following single particle states are available for interpretation: [622] $\frac{5}{2}^+$ and, with less probability, [624] $\frac{7}{2}^+$, [631] $\frac{3}{2}^+$ and [752] $\frac{5}{2}^-$. The assignment [752] $\frac{5}{2}^-$ can be ruled out, because the resulting multipolarities 330 keV-$M1$, 199 keV-$E1$ and 161 keV-$E1$ cannot explain the high intensity of the K X-rays in the 4820 keV alpha-gamma coincidence spectrum. The ratio of the reduced theoretical transition probabilities of the 161 keV and 199 keV gamma rays has the same value of 0.40 as for the interpretation [622] $\frac{5}{2}^+$. However, these lines would be strongly hindered $E1$ radiations, so that in addition to the more intense 330 keV line (as an unhindered $M1$ transition to the ground state) an unhindered $E2$ transition to the $(\frac{7}{2}, \frac{9}{2}^-)$ level should occur with an intensity comparable to that of the 199 keV line. Such a gamma ray was not observed. Likewise, the interpretation [631] $\frac{3}{2}^+$ can be ruled out because in addition to the 330 keV line – which should then be interpreted as $M1$ transition to the $(\frac{1}{2}, \frac{1}{2}^+)$ level – the two $M1$ transitions to the $(\frac{1}{2}, \frac{3}{2}^+)$ and $(\frac{1}{2}, \frac{5}{2}^+)$ level should exist. The reduced transition probabilities should behave as 0.5:0.4:0.1. These $M1$ transitions to the $(K=\frac{1}{2}^+)$ rotational band would be unhindered and they should be much more intense than the hindered 199 keV-$M1$ gamma ray (by a factor of about 500). Assuming the classification [624] $\frac{7}{2}^+$ the reduced transition probabilities of the $E1$ transition to the $(\frac{7}{2}, \frac{9}{2}^-)$ level and of the 330 keV-$E1$ transition to the ground state should behave as 0.286:1. These transitions would be hindered, the $M1$ transitions to the $(K=\frac{5}{2}^+)$ band with about 104 keV (which is then expected), 161 keV and 199 keV being unhindered. Thus the observed hindrance of the 330 keV gamma ray could possibly be explained by the interpretation [624] $\frac{7}{2}^+$ of the 333 keV level. However, because no indications were found for the above $E1$ transition to the $(\frac{7}{2}, \frac{9}{2}^-)$ level the orbital [624] $\frac{7}{2}^+$ must be ruled out, too.

The interpretation [622] $\frac{5}{2}^+$ can consistently account for the intensity ratios of the 161 keV, 199 keV and 330 keV transitions. Dipole transitions to levels of the $(K=\frac{1}{2}^+)$ rotational band are K-forbidden. In fact, such transitions were not observed. The only K-allowed dipole transitions are a hindered $M1$ to the $(\frac{5}{2}, \frac{7}{2}^+)$ level, a hindered $M1$ to the $(\frac{5}{2}, \frac{5}{2}^+)$ level and a much more retarded $E1$ transition to the ground state. These transitions very well fit the observed gamma lines at 161 keV, 199 keV and 330 keV. On the basis of the discussed branching ratios it seems safe to assign the quantum numbers [622] $\frac{5}{2}^+$ to the single particle level at 333 keV.

The levels at 368 keV, 424 keV, 470 keV and 526 keV can be arranged into a rotational band based on the [622] $\frac{5}{2}^+$ state. The rotational spacing factor of 5.2 fits well into the rotational spacing factors of the other three bands (cf. Table 7). The rotational levels should mainly be de-excited by transitions within this band because all direct dipole transitions to members of the other three rotational bands are either K-forbidden or hindered on account of the selection rules in the asymptotic quantum numbers. Indeed, no direct transition from the 368 keV level to another rotational band could be established. The 189 keV and 260 keV transitions which were ascribed to the $(\frac{5}{2}, \frac{9}{2}^+)$ level at 424 keV belong to the weakest gamma rays of Pu^{239}. The ratio of their reduced theoretical transition probabilities has the value $B(M1, 189\,\text{keV})/B(M1, 260\,\text{keV}) = 0.82$ and is not in contrast to the experiment (cf. Table 6).

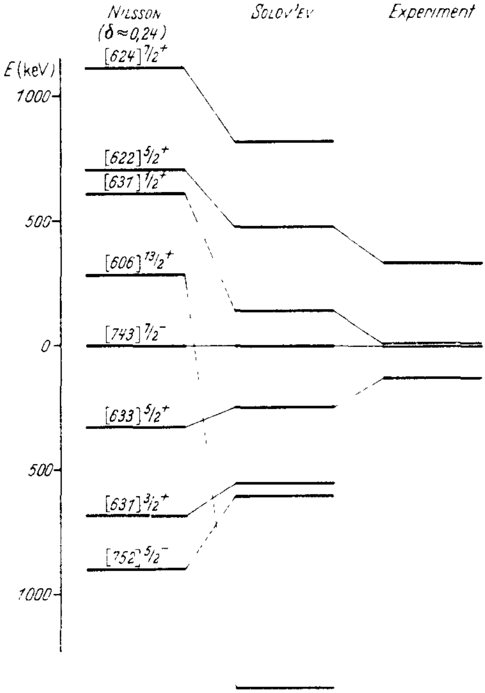

Fig 9 Comparison of the intrinsic levels predicted by the Nilsson model and the superfluid model of Solov'ev with the experimental results

Difficulties still exist concerning the relatively high intensity of the 4730 keV alpha group which can hardly be explained only by a $(\frac{5}{2}, \frac{9}{2}^+)$ level at 424 keV. Such an intensity step as observed between the neighbouring alpha lines and the 4730 keV alpha line does not appear anywhere else in the alpha spectrum. This fact may suggest a new single particle level at about 424 keV. In fact, Baranov et al. observed two alpha groups in this region and they assumed excited levels at 422 keV and 426 keV. The intense 380 keV transition to the $(\frac{7}{2}, \frac{9}{2}^-)$ level might be ascribed to such a single particle state at 424 keV. It can hardly be attributed to the $(\frac{5}{2}, \frac{9}{2}^+)$ rotational level, because no transition to the $(\frac{7}{2}, \frac{11}{2}^-)$ level could be observed. This transition should occur with a relatively strong intensity as is indicated by the theoretical ratio of the reduced transition probabilities $B(E1, \frac{9}{2} \rightarrow \frac{11}{2})/B(E1, \frac{9}{2} \rightarrow \frac{9}{2}) = 2.03$.

The 4685 keV alpha-gamma coincidence spectrum exhibits a line at 420 keV which is clearly observed in the total gamma ray spectrum. Most probably, this transition reaches the $(\frac{7}{2}, \frac{9}{2}^-)$ level at 46 keV. In interpreting the 470 keV level as $(\frac{5}{2}, \frac{11}{2}^+)$ rotational level there should be a transition to the $(\frac{7}{2}, \frac{11}{2}^-)$ level with a ratio of reduced transition probabilities of $B(E1, \frac{11}{2} \rightarrow \frac{11}{2})/B(E1, \frac{11}{2} \rightarrow \frac{9}{2}) = 8.3$ parallel to the 420 keV transition. In fact, the 4685 keV coincidence spectrum shows a gamma ray at 380 keV which is about three times more intense than the 420 keV line. However, it cannot be definitely derived from the present experiments whether this line is the transition already observed in the 4730 keV coincidence spectrum or a second 380 keV gamma ray. In the first case a transition between the 470 keV level and the possible single particle state at 424 keV should exist which would suggest the 470 keV level to be a rotational level based on the 424 keV single particle state. The 245 keV transition to the $(\frac{5}{2}, \frac{9}{2}^+)$ level at 229 keV observed in the gamma-gamma coincidence spectra is consistent with the interpretation of the 470 keV level as $(\frac{5}{2}, \frac{11}{2}^+)$ rotational state and, accordingly, would be a $M1$ radiation.

4.6. Level Density. Fig. 9 gives a comparison of the experimental intrinsic level excitation energies with the predictions of the Nilsson theory and the superfluid model. Obviously, the experimental results justify the modifications introduced by Solov'ev. The single particle levels found in U^{235} seem to point to an even higher level density than that calculated by Solov'ev.

The author wishes to thank Dr. W. Michaelis for his continued interest and guidance in this work.