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ELECTRON-CAPTURE DECAY OF ^{204}Po

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Introduction

The properties of light polonium isotopes of mass number less than 205 are not known well, owing to their relatively short lifetimes. These isotopes decay chiefly by electron capture, the alpha decay contribution being small.

^{204}Po was first identified by Karraker, Templeton, and Ghiorso[1] who reported a 3.8-h half-life and a 5.37 MeV alpha-particle energy. Further investigation led to the results summarized in Table 1. The branching ratio for alpha decay is 0.65%. In this work, a study was made of the electron capture transitions ^{204}Po → ^{204}Bi which permit identification of the unknown energy levels of the odd-odd ^{204}Bi nucleus. The only information about ^{204}Bi levels is due to Stoner[5], whose investigations of the alpha decay of ^{208}At demostrated the existence of a level of 120 ± 10 keV. This isotope seems to be of interest since it has one proton beyond the closed magic shell 82 and five neutron holes in magic shell 126. A great deal of experimental and theoretical work, with various methods of interpretation, has been done for near lead-208 nuclei. Prediction of the spin energies and level parities of spherical nuclei requires the use of theories taking into account quasi-particles and collective degrees of freedom.

However, for low-lying levels of nuclei in the vicinity of closed shells, as shown by Pryce /6/, the energy of a configuration of holes and particles can be taken as equal, in the first approximation, to the sum energies of the individual holes and particles in the effective field of the nucleus (zero-order energy). The short-range interaction of particular pairs of nucleons and holes can be taken into account by using the semi-empirical two-particle interaction parameters given by Altburger and Pryce /7/ (first-order energy). This method has been employed successfully to explain the established levels of ^{204}Pb /8//9/, ^{205}Pb /6/, ^{206}Bi and ^{207}Bi /10/. Kim and Rasmussen performed the calculations for ^{210}Bi /11/ and for ^{208}Tl and ^{208}Bi /12/. They obtained a good fit to the low excited states, by using the j-j coupling shell model and a residual force, including the tensor force. Light isotopes of lead were analyzed by Kisslinger and Sorensen /13/.

In the calculations, the neutron hole states of Pb isotopes in the mass number range 195-207 were considered to interact with a pairing force and with a $P_2(\cos\theta)$ long-range force. The results predict the ground state spins and correct excited level positions in nearly all odd-A isotopes.

As we move away from the closed-shell configuration, the increasing number of degrees of freedom leads to ever more pronounced generation of collective effects.

In considering the high-energy levels, one must take into account the possible excitation of collective states as well as particle-core coupling in odd-mass nuclei; the latter effect has been discussed by e.g. de-Shalit /14/.

In the lead region, a doublet in ^{207}Pb and a multiplet in ^{209}Bi /15/, can be explained by the coupling of particular single particle states to the octupole 2.62 ^{208}Pb state. Recently, Nelson Stein, Whitten, and Bromley /16/, by using inelastic proton scattering in the proton energy range 12-20 MeV, have demonstrated the existence of high-energy multiplets in ^{207}Pb corresponding to particle-plus-core states based on collective 2+, 4+, and 3-states in ^{208}Pb.

Experiment

The isotope ^{204}Po was obtained from the alpha decay of ^{208}Em produced by the reaction $^{197}Au(^{15}N, 4n)^{208}Em$. The short half-life of the ^{208}Em (23 min.) and the relatively large alpha branching (20%) permits effective production of ^{204}Po in this way. The cross section for this reaction has not been measured but data is available for the reaction $^{197}Au(^{14}N, xn)^{211-x}Em$ /17/. The maximum of the excitation curve for the emission of 4 neutrons reaches a value of 120 mb at an excitation energy of 52 MeV. The width of the excitation curve peaks for the emission of 4,5, and 6 neutrons is of the same order of magnitude as the spacing between peaks (≈ 10 MeV). It was thus reckoned that considerable quantities of ^{207}Em and ^{208}Em could be produced even if the energy of the bombarding ^{15}N ions corresponded to the maximum for the emission of 4 neutrons.

Taking into account the successive captures and alpha decays **Em → At → Po** and **Em → Po**, one had to expect the presence of polonium isotopes 202 to 208 in the source. Gold foil 10 μ thick was irradiated with a ^{15}N ion beam having an energy of 80 MeV and intensity of 1 μ A. The irradiation time was 5 hours. After irradiation the target was subjected to chemical operation.

Polonium was separated from irradiated gold by coprecipitation with elementary tellurium, formed in situ by reduction with stannous chloride. The precipitate was dissolved in a 50 per cent nitric acid, and after the addition of 6N hydrochloric acid tellurium was eliminated by precipitation with sulfur dioxide.

The polonium then was deposited spontaneously onto silver (or onto plâtinum) from 0.1 N hydrochloric acid at 70-80° C.

The source thus obtained could, in principle, be used for measuring gamma and alpha spectra. However, better results for alpha spectra were obtained by absorbing polonium on a platinum foil (thinner source) although the extraction efficiency was lower in this case. Figure 1 presents the alpha spectrum recorded by a semiconductor detector with a resolution of 19 keV, 170 minutes after irradiation was completed. The broadening of all the polonium peaks to about 28 keV is due to the source thickness. Polonium isotopes of mass number in the range of 202 to 207 were identified in this spectrum. As can be seen, the ^{204}Po peak is predominant with respect to the others. The peak corresponding to ^{208}Po, whose specific activity is low, presumably coincides with the ^{207}Po peak and the 5.22 MeV peak is the sum of the values recorded for ^{206}Po (5.218 MeV) and ^{205}Po (5.2 MeV). If account is taken of the short life-times of the identified polonium isotopes and of their quantities, it is seen that only ^{207}Po with a life-time of 5.7 h could hinder measurements. The intensity of this peak is smaller by a factor of 27 than that of ^{204}Po , but in view of the fact that the branching α/EC is 0.014% for ^{207}Po /19/ while it is 0.63% for ^{205}Po /20/, the electron capture contribution from ^{207}Po becomes half that from ^{204}Po and increases with time. It has turned out, however, that the presence of ^{207}Po , did not hinder the measurements but, in fact, proved helpful in checking the energy scale calibration.

The gamma spectra were measured by a germanium detector with volume 8.5 cm^3, and resolution 5 keV. Pulses from the amplifier were fed to a 4096 channel analyser. During the measurements the end channels of the analyser recorded a line from a generator with a random time distribution and this made it possible to estimate the effect of the dead time on the recording effeciency in the individual series in which the intensity varied. The maximum correction between the series separated most in time was 3.5% . For the germanium detector measurements of efficiency were performed in the energy range up to 1.5. MeV.

The spectrometer was calibrated by means of 12 lines from standard sources. Additional verification of the calibration was provided by gamma lines from the decay of ^{207}Po and ^{204}Bi occurring in the measured spectra. The intensive lines og ^{207}Po — 249.6, 742.7, 992.6 and 1148.8 keV and of ^{204}Bi — 375.0 and 899.2 keV for which the intensity ratios are well known /21, 22/, were furthermore used to to check the efficiency of the detector. The ratios of the intensities of these lines, as determined in the course of this work, do not differ by more than 2% from the data in Refs. /21,22/. The cut-off of the electronic circuit did not permit gamma-ray quanta of energy less than 60 keV to be recorded. The results obtained from the measurements come from two irradiations. The lines in the range of energies from 70 to 270 keV come from the first irradiation whereas the 270–1400 range comes from the second. The 270 keV, peak constituted the basis for normalization of both series.

Recording of spectra in the first series five times at half-hour intervals and three times about 2 hour intervals, enabled the lifetimes of the individual lines to be evaluated and their genetic origin to be determined.

Results

Figure 2 presents the gamma spectra of two consecutive measurements made at an interval of 2.05 h (2a – range of 0– 1000 channels, 2b – 1000 – 2000 channels) . One can easily see rapidly vanishing peaks and rising peaks, mainly of ^{204}Bi , a decay product of ^{204}Po : e.g. 290, 375, 899 keV. In addition to the lines indicated in Fig.2, several lines of low intensity were recorded in the high-energy region. Owing to the low efficiency of the germanium detector in this energy range, a very small over-all number of pulses in these peaks was recorded and their lifetimes could be evaluated only roughly.

Comparison of the energies of the individual peaks, determined in the individual series of measurements, indicated that the operation of the system was very stable. In general, the peak energies could be found to within 0.5–0.7 keV and at the end of the spectrum ($E_\gamma \geq 900$ keV) where the number of calibration points was smaller, to within 0.8 – 1.0 keV. The first two lines, 76 and 88 keV, correspond to unresolved X-rays: K_α ,and K_β of bismuth with small contributions by lead from bismuth decay. Knowing the intensity ratios of K_{α_1}, K_{α_2} , K_{β_1} , K_{β_2} , /23/, we obtain for bismuth a computed value of $I_{K_\alpha}/I_{K_\beta} = 3.38$, while our experimental data give 3.33.

Table 2 lists lines identified on the basis of measurements of lifetimes as lines which unquestionably come from the decay of ^{204}Po; the energies and relative intensities of the lines, as computed with account for the efficiency of the germanium detector, are given.

Construction of the energy-level diagram of ^{204}Bi was started from the 120 keV level which was determined by Stoner in investigations of the alpha decay of ^{208}At /5/. Two sums can be obtained by starting from the 120 keV level: 120 + 306 = 426 and 120+ +762 = 882. The 882 keV level would occur in three further combinations: 882+ 247 = 1129; 882 + 136 = 1018; 882 – 204 = 678. If we assume that the 678 keV and 1129 keV levels exist, we have a 451 keV transition between them: 678 + 451 = 1129. Proceeding in turn from the 678 keV level, we can confirm the existence of a weak 1720 keV transition which undoubtedly comes from the decay of ^{204}Po : 678 + 1042 = 1720. Two strong gamma lines, 535 and 270 keV, still remain. We have 535 = 317 + 218, whereas 270 does not appear in any of the combinations of sums or differences.

The tentative decay scheme of ^{204}Bi, as constructed on the basis of the presented relations, is shown in Fig.3. Coincident measurements are undoubtedly necessary for establishing more reliably the decay scheme, and measurements of the conversion electron spectra are needed for determining the multipole nature of the transitions; the latter would, in turn, permit the spins and particles of the levels to be established. An attempt can be made to obtain some information by considering the spin and parities of the possible configurations of "valence" nucleons and holes.

If we take the order of hole levels to be $p_{1/2}$, $f_{5/2}$, $p_{3/2}$, $i_{13/2}$ and the energies in those states to be 0.00, 0.32, 0.57, and 1.63 MeV, respectively /6/, whereas for protons in the states $h_{9/2}$, $f_{7/2}$ and $i_{13/2}$ energies of 0.00, 0.90 and 1.56 MeV, respectively, we can present several possible configurations of these spins and parities and zero-order energies in the form of a table (Table 3).

Detailed calculation of the weights of $(f^2_{5/2})$ configuration in ^{205}Pb shows /6/ that the coupling to I =0 is dominant (98%) and thus the configuration $(p^2_{1/2}\, f^2_{5/2})_{I=0}\ f_{5/2}$ should predominate. The ground states of ^{203}Pb has a spin of 5/2 /24/ which would confirm this hypothesis whereas the first and second excited states of spin 1/2-, and 3/2- would presumably correspond to the configuration

$(f^{4}_{4/2})_{I=0}\ p_{1/2}$ and $(p^{2}_{1/2}\ f^{2}_{5/2})_{I=0}\ p_{3/2}$.

In the computations summarized in the table, only the confuguration $(f^{2}_{5/2})_{I=0}$ was also taken into account. Greater clarity of the possible spins has been obtained at the price of seemingly small error. The spins of value $J = j_1 + j_2 - 1$ for $j_2 \geq 3/2$ (weak-coupling rule) /25/, have been underlined.

Discussion

The gamma lines identified in our experiment may provide a basis for, at the most, the inital speculations concerning the energy-level scheme of ^{204}Bi . Inasmuch the spins of ^{204}Po (0 +) and ^{204}Bi (6 +) differ greatly, the decay ought to populate excited states of ^{204}Bi (available transition energy 2.168 MeV /26/) and a complex gamma spectrum could be expected.

The compilation of possible spins and particles of the individual configurations (table 3) indicates that not many levels may be accessible for E.C. transitions, even if account is taken of the first and second forbidden transitions. The configurations involving the conversion of an $h_{9/2}$ proton into an $f_{5/2}$ neutron are most likely.

The spins and parities of the individual configurations point to the possibility of many transitions of multipolarity M1 and E2 in cascades to the ground state.

If a configuration with state $i_{13/2}$ was realized in the excited state of ^{204}Bi , one could expect isomerism resulting from the transition of the $i_{13/2} \rightarrow f_{5/2}$ type, just as in the case of lead isotopes. There may be reservations as to the tentative decay scheme presented. It has been constructed on the basis of the 120 keV level known from alpha decay. However, low-lying ^{206}Bi levels at 83 and 167 keV, respectively, found in the alpha decay of ^{210}At /27/, are not involved in the electron capture of ^{206}Po and this is rather puzzling. A drawback of the hypothetical scheme presented is that there are no levels in the interval from 1129 to 1720 keV. In the construction of a level scheme, by taking the sums and differences of the individual pairs of gamma lines, one could loose those levels of transition which form a cascade only and from which there is no direct transition to the ground state. Such a possibility refers in particular to high-energy levels.

Perhaps existing, but undetected levels would permit a different assignment of, for instance, the strong 270 keV line which does not occur in any combination of sums and differences.

The relatively high accuracy of our measurements ($\Delta E \approx 0.1\%$) has encouraged us to make the first attempt at formulating the decay scheme.

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E_α MeV	$T_{1/2}$ hr	Reaction	Ref.
5,37	3.8	Bi + p	1
5.370	3.8	Bi + p	2
5,38	3-4	Pt + $^{12,13}C$	3
5.39	3.6	Pt + ^{12}C	4

Table 1. Alpha-particle energies, and half-lives of ^{204}Po as measured by different research teams.

No	E_γ	I	No	E_γ	I	No	E_γ	I
1	120	3.3	8	317	13.3	15	882	74.0
2	136	27.1	9	426	7.7	16	1018	50
3	204	12.8	10	438	5.7	17	1042	25.6
4	218	4.4	11	451	5.0	18	1130	0.8
5	247	5.0	12	535	32.0	19	1720	0.8
6	270	68.7	13	678	21.5			
7	306	11.0	14	762	28.6			

Table 2. Gamma transitions, and their intensities in the decay of ^{204}Po.

Configuration	spin values	zero-order energy	π
$(p^2_{1/2}f^2_{5/2})f_{5/2}h_{9/2}$	2,3,4,5,$\underline{6}$,7	0,00	+
$(p^2_{1/2}f^2_{5/2})p_{3/2}h_{9/2}$	3,4,$\underline{5}$,6	0,32	+
$(f^4_{5/2})p_{1/2}h_{9/2}$	4,5	0,57	+
$p_{1/2}(f^2_{5/2})f_{5/2}p_{3/2}h_{9/2}$	0,1,$(2)^4$,$(3)^6$,$(4)^7$,$(5)^7$,$(6)^6$,$(7)^4$,$(8)^3$,9,	0,89	+
$(p^2_{1/2}f_{5/2})^2f_{5/2}f_{7/2}$	1,2,3,4,$\underline{5}$,6	0,90	+
$(p^2_{1/2}f^2_{5/2})i_{13/2}h_{9/2}$	2,3,4,5,6,7,8,9,$\underline{10}$,11	1,06	−
$(f^4_{5/2})f_{5/2}h_{9/2}$	2,3,4,5,$\underline{6}$,7	1,14	+
$(f^2_{5/2}p^2_{3/2})p_{1/2}h_{9/2}$	4,5	1,22	+
$(p^2_{1/2}f^2_{5/2})p_{3/2}f_{7/2}$	2,3,$\underline{4}$,5	1,23	+
$(f^4_{5/2})p_{3/2}h_{9/2}$	3,4,$\underline{5}$,6	1,46	+
$(f^4_{5/2})p_{1/2}f_{7/2}$	3,4	1,47	+
$p_{1/2}(f^2_{5/2})f_{5/2}p_{3/2}f_{7/2}$	$(0)^2$ $(1)^5$ $(2)^7$ $(3)^8$ $(4)^8$ $(5)^7$ $(6)^5$ $(7)^3$ 8	1,79	+

Table 3. Zero-order energies of ^{204}Bi configuration for five neutron holes, and one proton in $h_{9/2}$ and $f_{7/2}$ states. The holes configuration $(f^2_{5/2})_{I=0}$ was taken as dominant.

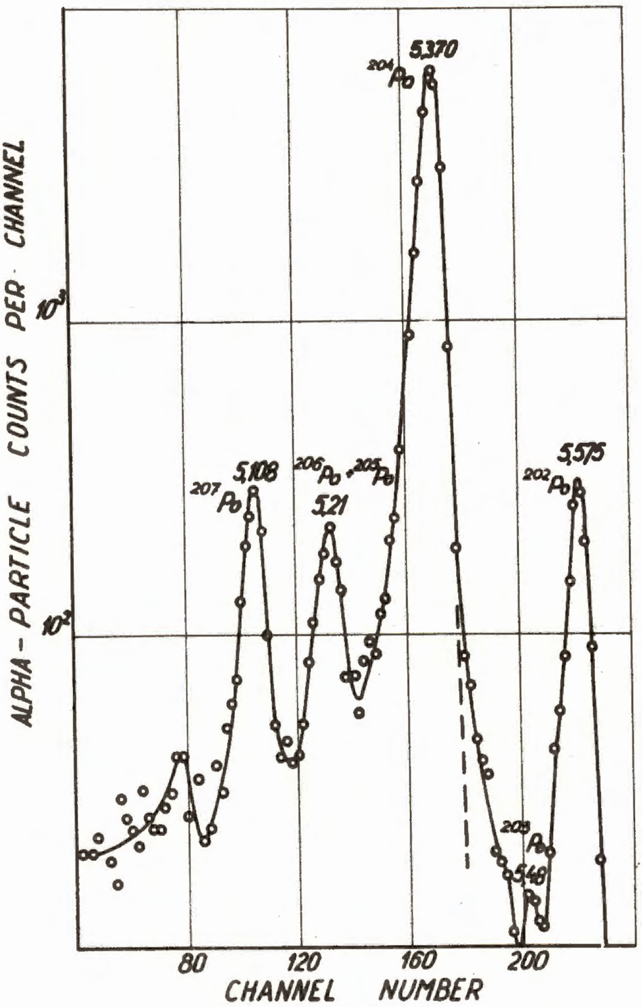

Fig.1. Alpha-particle spectrum of light polonium isotopes registered 170 min. after the irradiation of Au target with 80 MeV ^{15}N ions. The polonium was extracted onto platinium foil.

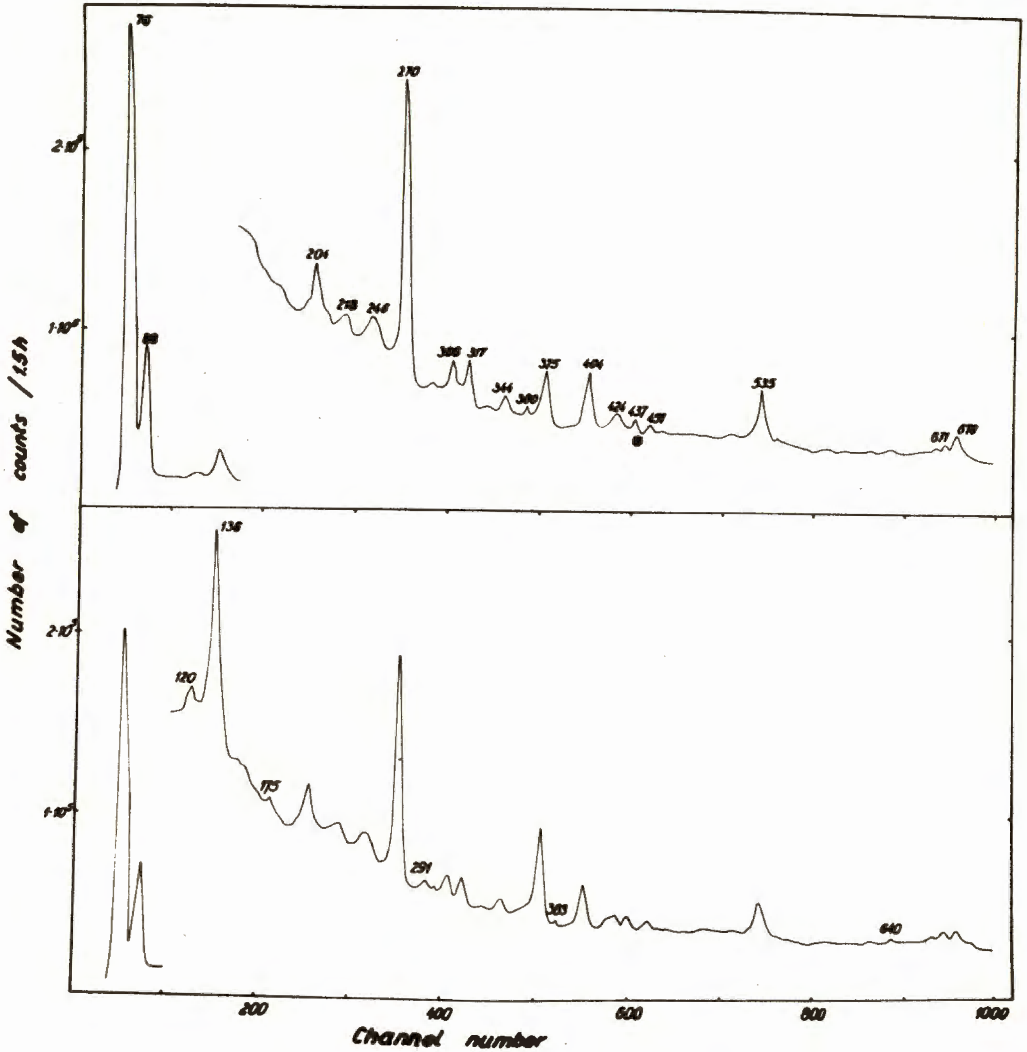

Fig.2a. Low energy part of the gamma-ray spectrum as measured with germanium detecror. The registration time was 1.5 h, and the interval between two series 2.05 h.

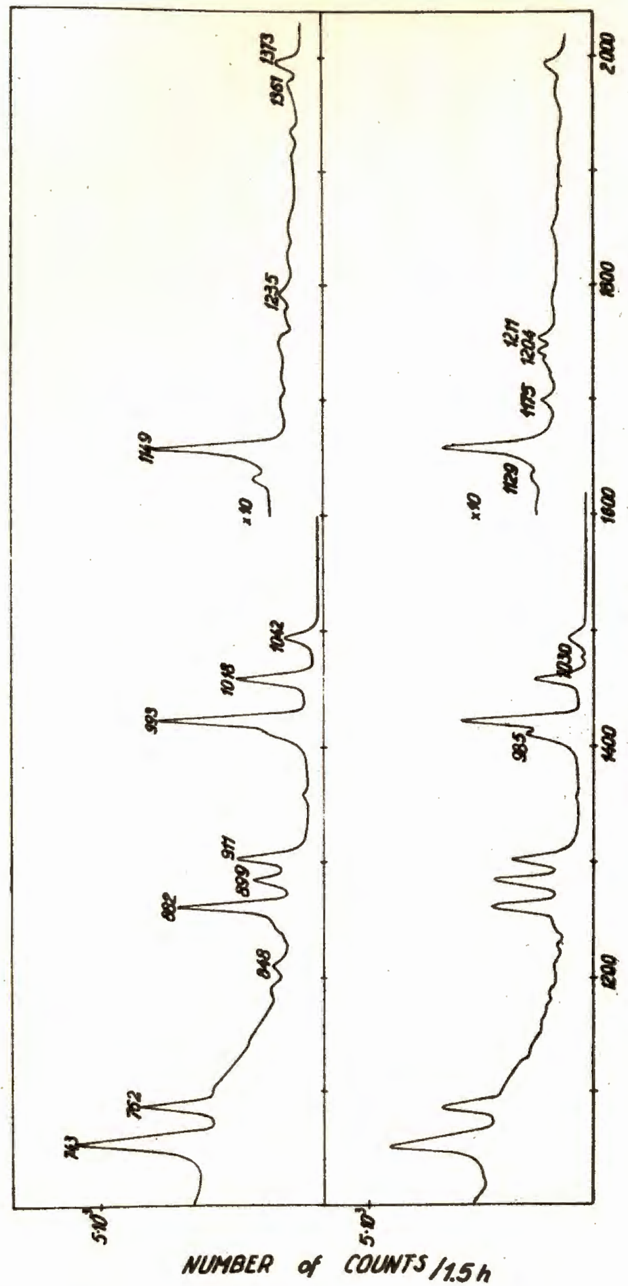

Fig.2b. High energy part of the gamma-ray spectrum.

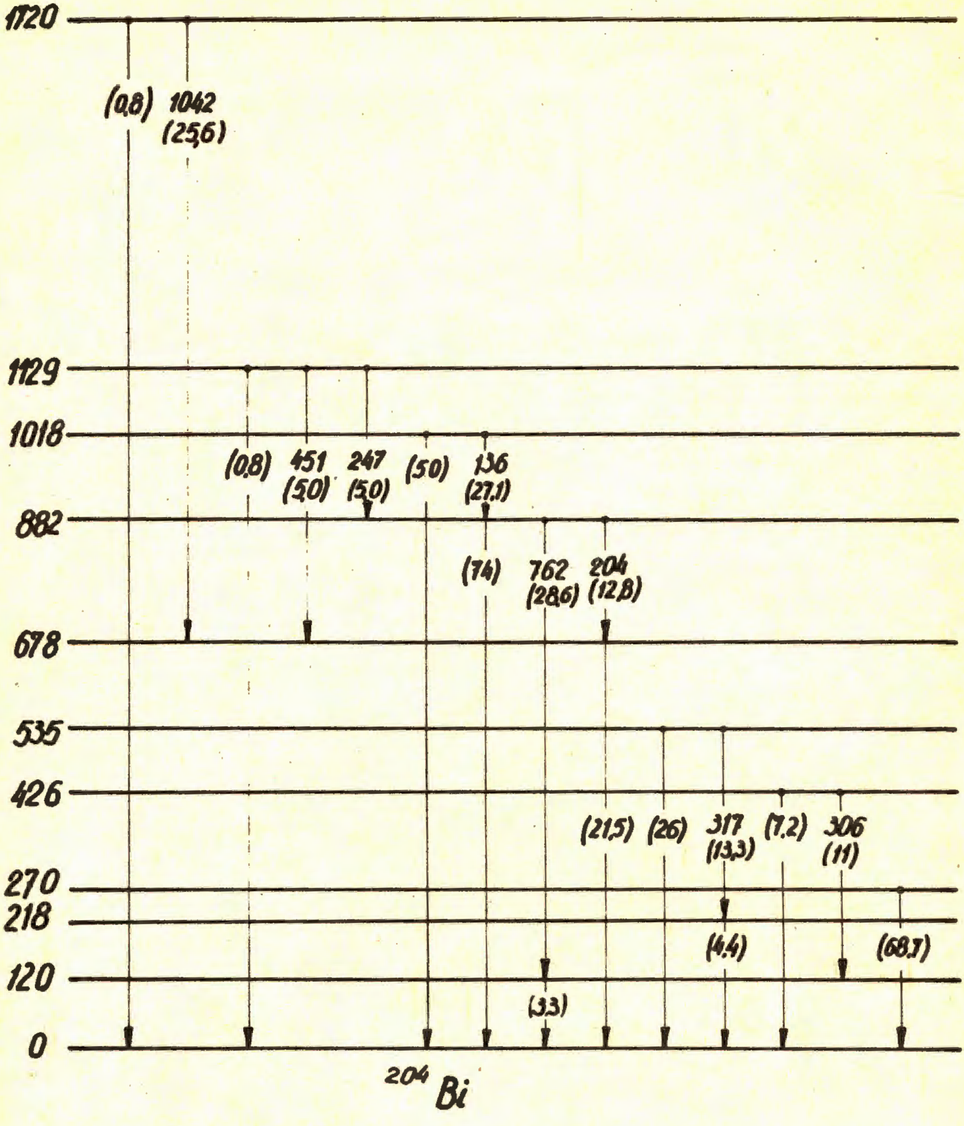

Fig. 3. Tentative decay scheme of ^{204}Po . Transition intensities are given in brackets.