ENERGY SPECTRA OF ALPHA PARTICLES FROM THE LONG-LIVED ISOTOPES Th^{232} *AND* U^{238}

G. E. KOCHAROV, A. P. KOMAR, and G. A. KOROLEV

Leningrad Physico-Technical Institute, Academy of Sciences, U.S.S.R.

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The alpha spectra of Th^{232} and U^{238} have been investigated by means of an ionization chamber with a grid. The energies and intensities of the transitions to the 2^+ and 4^+ states of the daughter nuclei were determined.

INTRODUCTION

INVESTIGATION of the fine structure in alpha-decay spectra opens a way for the study of weakly-excited states of heavy nuclei, and makes it possible to determine some of the parameters associated with their nuclear structure. The study of fine structure in alpha-decay spectra is carried out chiefly by means of magnetic spectrometers. However, the low aperture ratio (product of the source area by the solid angle used) of the latter does not permit their use in the study of fine structure in the alpha-decay spectra of long-lived isotopes such as Th^{232} and U^{238}, for example.

To solve this problem it is convenient to use ionization chambers whose aperture ratio is several orders of magnitude greater than that of a magnetic spectrometer. In spite of their high aperture ratio, however, ionization chambers are inferior to magnetic spectrometers in energy resolving power. Until recently, the half-width of alpha spectral lines from ionization chambers was of the order of 50 kev because of their poor energy resolution. But in many cases the separation between alpha lines is found to be less than 50 kev, and it is therefore impossible to resolve such lines completely. Hence any improvement in the resolving power of ionization chambers would be of great importance. During 1955 and 1956 a gridded ionization chamber was developed in our laboratories with a half-width of 30 kev for alpha lines.[1-4] The resolving power thus attained proved to be sufficient for a study of the fine structure of the alpha spectra of Th^{232} and U^{238}.

In studying the alpha transitions to excited levels of the daughter nuclei, particular attention had to be paid to the stability of the apparatus, since the measurements were carried out uninterruptedly for periods of several days or more. For instance, in studying the alpha transition to the 4^+ level of Ra^{228} the measurements were taken for 90 hours. During this time the drift of the apparatus used was less than 0.1% per day.

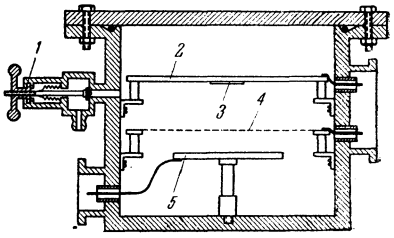

FIG. 1. Diagram of gridded ionization chamber. 1 – valve; 2 – high-voltage electrode; 3 – alpha-particle source; 4 – grid; 5 – collector electrode.

1. DESCRIPTION OF THE APPARATUS

1. The Ionization Chamber and Its Operation

A diagram of the chamber is shown in Fig. 1. The source of alpha particles is located inside the high-voltage electrode, and consists of a brass disk 220 mm in diameter and 4 mm thick. The grid is made of nichrome wires 0.1 mm in diameter welded to a ring of stainless steel. The distance between adjacent wires is 1.5 mm. The grid is 50 mm away from the high-voltage electrode and 25 mm away from the collector electrode. The potentials of the ion chamber electrodes were chosen so as to exclude the possibility of ion recombination or of electron capture by the grid. The chamber was filled with a mixture of 97% argon and 3% methane, to a pressure of 1.8 atmos. The pulses from the collector were amplified in a low-noise preamplifier before being fed into the main amplifier, which had a gain of 10^3 (see Fig. 2). 6Zh1P tubes were used in the preamplifier. Under optimum conditions the transconductance of the first tube was about 3000 μmho, with a grid current of 2×10^{-11} amp. A model-100 amplifier[5] was used for the main amplification. The time constants of the integrating and differentiating circuits were both equal to 15

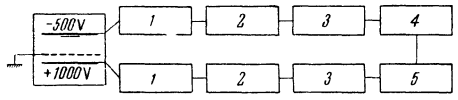

FIG. 2. Block diagram of the ionization alpha-spectrometer. 1 – preamplifiers; 2 – amplifiers; 3 – limiters; 4 – discriminator; 5 – 28 channel analyzer.

microseconds. To reduce the width of the analyzer channels, the output pulses from the amplifier were fed through a limiting amplifier.[6] From the output of this limiter the pulses went to a 28-channel amplitude analyzer. By varying the gain of the limiting amplifier, it was possible to change the effective width of the analyzer channels. The minimum channel width used in the present investigation was 0.1% of the alpha-particle energy. The pulses originating at the high-voltage electrode were used to provide electrical collimation.[3,4]

2. Energy Resolution of a Gridded Ionization Chamber

The energy resolving power of a gridded ionization chamber depends on the thickness of the source, the purity of the gas, the background electrical noise, and the geometry and operating conditions of the chamber. We shall now consider the most serious causes of straggling in the pulse amplitudes.

Fluctuations in the number of ion pairs, and also electrical noise, lead to a Gaussian distribution of pulse amplitudes; but the source thickness, lack of perfect screening by the grid, etc., lead to distributions which are not Gaussian. In spite of this, it is customary to use some value of effective mean-square deviation as a rough guide to the relative effects of each of the above-mentioned factors on the half-width of an alpha line. It will be understood that such estimates are not necessarily mathematically rigorous.

a. Ionization fluctuations. The mean square fluctuation in the number of ion pairs is equal to[7] $\Delta N^2 = FE/W$, where E is the alpha-particle energy, W is the mean energy required to form a single ion pair, and F is a parameter depending on the nature of the gas. According to Fano,[7] F is $\frac{1}{3}$ for argon. The corresponding value of the mean-square deviation for alpha particles from U^{238} is $s_i = 6.3$ kev.

b. Electrical noise. The signal-to-noise ratio at the amplifier output depends on the characteristics of the first amplifier tube as well as on the correct choice of the pass band. To obtain the maximum signal-to-noise ratio for a given amplifier bandwidth, the tubes must have high transconductance and low grid current. By a suitable selection of the first amplifier tube and its operating potentials, it was possible to reduce the mean-square value of electrical noise to a value equivalent to a spread of 6.8 kev.[1]

c. Effect of source thickness on resolution. In studies of long-lived isotopes the use of thick particle sources is unavoidable. Particles emitted from the deeper layers of the source, and particles coming off at large angles to the normal at the surface of the source, lose part of their energy within the source itself. The result is an increased half-width of the alpha line and the appearance of a "tail" on the low-energy side. The presence of this large "tail" in a pulse height distribution makes it difficult to study the fine-structure components. It is therefore necessary to apply some collimation. Mechanical collimators are ordinarily used for this purpose.[6,8] However, they suffer from a number of disadvantages: (1) some of the electrons produced by the alpha particles are lost because of the weakening of the electrical field in the collimator aperture, and (2) the geometry of the apparatus is seriously reduced. Another essential drawback is the difficulty of fabricating highly uniform collimators.

In our work we used the method of electrical collimation. To provide electrical collimation we made use of the pulses produced at the high-voltage electrode. It is well known that the differential distribution of the pulses is rectangular.[9] At the same time, the maximum amplitude V_{max} corresponds to a particle emitted parallel to the electrode, while the minimum amplitude V_{min} corresponds to one emitted perpendicularly. It is obvious that to achieve collimation it is necessary to record the number of collector pulses which coincide with the high-voltage electrode pulses whose amplitudes lie in the interval from V_{min} to $V_{min} + \Delta V$. By changing the magnitude of ΔV, the degree of collimation can be altered. In realizing electrical collimation we used an ordinary single-channel differential amplitude discriminator. The degree of collimation depends on the alpha-particle energy, thus distorting the natural intensity ratio. At the same time, it proved to be easy to allow for the intensity ratio.*

Taking into account the degree of collimation and the effect of source thickness on the shape of the alpha spectrum,[10] we arrive at an rms value $s_S \approx 4$ kev for U^{238} alpha particles.

d. Electron Trapping by Electronegative Impurities. The presence of electronegative gas impuri-

*G. E. Kocharov, Diploma thesis, Phys. Tech. Inst. Acad. Sci. U.S.S.R. (1956).

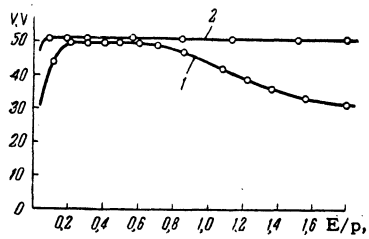

FIG. 3. Height of the collector electrode pulse as a function of the ratio E/p. 1 – A + 0.05% O_2; 2 – A + 0.05% O_2 + 3% CH_4.

ties in the ionization chamber leads to a loss of electrons moving toward the collector electrode and reduces the energy resolving power. The most active electronegative gas is oxygen. Gases are usually rid of oxygen by molten sodium or calcium. In a purified gas, for example argon, the electron drift velocity is small,[11] with the result that the resolution in time and energy is impaired (see item e). In this connection, it is of great practical value to use mixtures of gases. Facchini and Malvicini[12] have shown that the addition of a small amount of nitrogen to the argon prevents this trapping of electrons. The mixture of A and CH_4 which we have used behaves in the same way as a mixture of A and N_2. Figure 3 shows the experimental relationship which we observed between the pulse height and E/p for A + 0.05% O_2 and A + 0.05% O_2 + 3% CH_4. It is evident from these curves that the mixture of argon and methane is insensitive to oxygen impurities. In addition, the electron drift velocity is considerably increased in such a mixture,[11] and the total number of ion pairs formed by an alpha particle is also increased. The addition of 3% of CH_4 considerably improves the amplitude characteristics of the mixture.

e. Lack of Grid Screening. The grid is put into the chamber to suppress any disturbance of the collector electrode which might be induced by the positive ions. The inefficiency of the grid shielding can be determined from the size of the charge induced on the collector electrode by the ions formed in the space between the grid and the high-voltage electrode. As a measure of grid inefficiency we introduce the parameter $\sigma = dE_2/dE_1$, calculated from the formula[13]

$$\sigma = \frac{c}{2\pi b} \ln \frac{c}{2\pi r}, \qquad (1)$$

where c is the distance between grid wires, r is the wire radius, E_1 and E_2 are the electric field strengths in the regions between the grid and the high-voltage electrode and between the grid and the collector respectively, and b is the distance between the grid and the collector.

The grid inefficiency σ can be reduced by increasing the distance between the grid and the collector and the size of the wires, or by decreasing the separation between the grid wires. In so doing, it is essential to keep in mind that the electrons can settle on the grid wires.[13]

Incomplete grid screening makes the collector pulse amplitude depend on the angle at which the alpha particle is emitted, thus reducing the energy resolution. Bunemann et al[13] derived an analytical expression for the rms deviation

$$s = \sigma x E / 2f [d + l(1 - \sigma)], \qquad (2)$$

where x is the distance of the center of gravity of the ions along the alpha-particle track, E is the alpha-particle energy, d is the distance between the high-voltage electrode and the grid, l is a parameter depending on the grid geometry, and f is the degree of collimation.

The experimentally measured value of $\sigma = 0.014$ agrees with the calculated value.[13] Substituting all the necessary values into Eq. (2) we get $s \approx 3$ kev.

f. Effect of Different Collector-pulse Rise Times on Energy Resolution. The pulse rise time at the collector electrode depends on the angle of emission of the alpha particle, and is equal to

$$t = b/w_2 + R\cos\vartheta/w_1, \qquad (3)$$

where R is the range of the alpha particles in the chamber gas; w_1 and w_2 are the drift velocities in the regions between the high-voltage electrode and grid, and the collector and grid, respectively; and ϑ is the angle between the alpha track and the normal to the plane of the source.

During the process of amplification the pulses go through differentiation and integration networks, in which the output pulse amplitudes will depend upon the rise time. The dependence of the rise time on the angle impairs the energy resolving power. The use of mixed gases, A + 3% CH_4, to give a high drift velocity, together with the use of collimation, makes possible a considerable reduction in the straggling of pulse heights due to this effect. Using measured values of electron drift velocities in mixtures of argon and 3% methane,[11] and taking into account the degree of collimation, it is possible to estimate the effect of various pulse rise times on the resolution. The corresponding variance does not exceed 1 kev.

In addition to the above factors, the resolution also depends on the stability of the apparatus, etc. Hence the total rms variance, calculated from the formula $s = (\Sigma s_i^2)^{1/2}$, is equal to 11 kev, corresponding to a half-width of 26 kev in the curves.

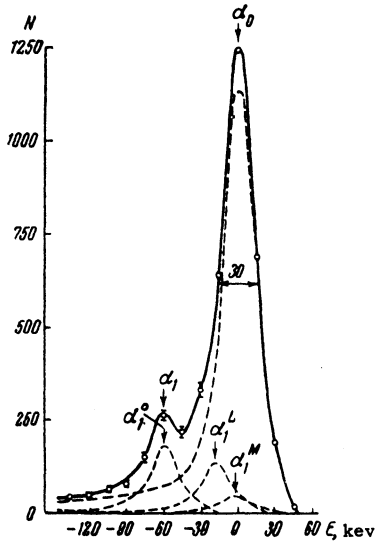

FIG. 4. Alpha particle spectrum of Th^{234}, taken over 25 hours. Half-width of alpha lines 30 kev. The resolution into components is shown dotted. α_0 is the fundamental group of alpha particles; α_1 is the group corresponding to transition to the 2^+ level of the daughter Ra^{228} nucleus. Half of the alpha of group α_1 are accompanied by conversion electrons from the L and M shells which cause excess ionization (groups α_1^L and α_1^M); α_1^0 corresponds to the cases where these electrons do not enter the working space of the chamber.

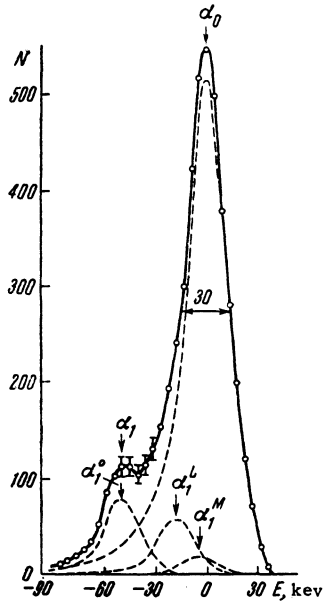

FIG. 5. Alpha particle spectrum of U^{238}, taken over 1.5 hours. Half-width of the lines is 30 kev. Resolution into components is shown dotted. Channel width 4.5 kev.

This is not significantly different from the experimental value of 30 kev.

2. RESULTS OF THE MEASUREMENTS

When a short-range alpha particle is emitted, the atom is left in an excited state. The transition to the ground state is accompanied by the emission of conversion electrons or gamma rays. Following the conversion electrons, the atom emits x-rays and Auger electrons. All these radiations which accompany the alpha-decay produce additional ionization in the chamber gas, thus increasing the ionization due to the short-range particle itself. The major portion of the additional ionization is due to the conversion electrons; the other radiations produce minor amounts of ionization which can be neglected. The working space of the chamber receives half of the conversion electrons. As a result, half of the pulses V_α from the alpha-particle fine structure lines coincide with pulses* from conversion electrons, V_e. This gives a total pulse equivalent to $V_\alpha + V_e$, corresponding to a group of alpha particles with an energy $E = E_\alpha + E_e$. The other half of the pulses register like pulses of alpha particles with the correct energy E_α. In the resulting pulse height distribution, a single group of alpha particles shows up at several different values of energy. This complicates the experimental determination of energies and intensities of alpha transitions. In working out the experimental results, therefore, account must be taken of the spectral distortion due to the conversion electrons.

*The ranges of the conversion electrons are all within the chamber working space.

1. Alpha Transitions to the First Excited Levels of the Daughter Nuclei Ra^{228} and Th^{234}

The alpha spectrum of Th^{232} is shown in Fig. 4. The measurement time was 25 hours. The stability and resolution were sufficiently good to permit the resolution of a well-developed peak α_1, corresponding to transitions to the first excited state of the daughter nucleus Ra^{228}. The distortion due to conversion electrons was taken into account in resolving the curve into its component lines. The intensity of the line α_1 is 23 ± 3%. The energy of the first level is 60 ± 5 kev. This value is in good agreement with measurements made on conversion electrons.[14]

We also succeeded in resolving the α_1 group of particles in the U^{238} spectrum (Fig. 5). The energy of the corresponding level is 48 ± 5 kev, and the intensity of the transition is 23 ± 4%.

It should be mentioned that after our measurements had been completed,[2] we learned that the α_1 group of U^{238} had also been obtained by Valladas,[15] using the coincidences of alpha particles with x-rays. He found the intensity of the transition to be about 20%.

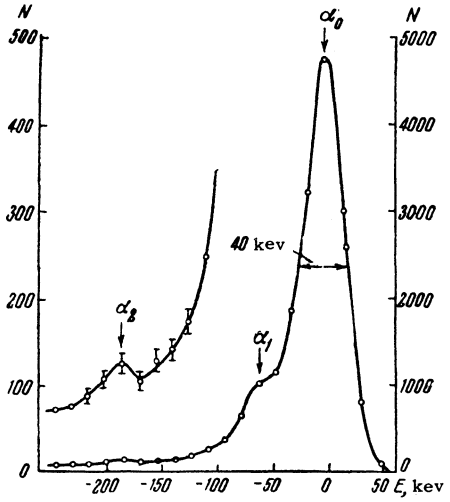

FIG. 6. Alpha particle energy spectrum of Th^{232}, taken over 90 hours. α_1 is the corresponding transition to the 2^+ level of the daughter nucleus. α_2 is apparently a group of alphas corresponding to a transition to the 4^+ level.

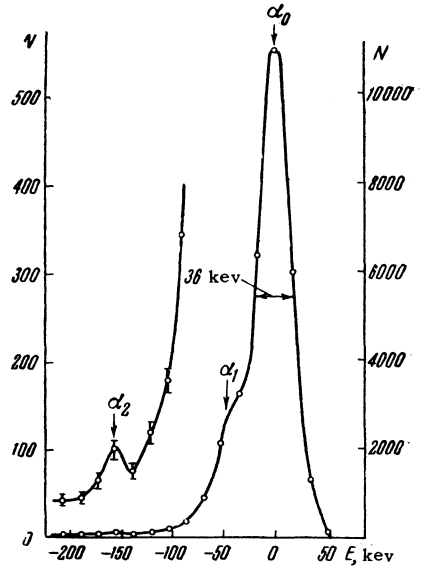

FIG. 7. Alpha energy spectrum of U^{238}, taken over 30 hours. The group α_2 obviously corresponds to a transition to the 4^+ level of the daughter nucleus Th^{234}.

2. Transitions to the Second Levels of the Daughter Nuclei Ra^{228} and Th^{234}

Up to the present time no data have been available on the second excited level of Ra^{228}, apparently because of the difficulties of studying the alpha spectra of nuclei with long half-lives.

The high aperture ratio and good energy resolution of our ionization chamber made it possible to observe an alpha-transition to the second excited level of the daughter nucleus in the spectrum of Th^{232} (Fig. 6). The time required for the measurement was 90 hours. From an analysis of the experimental curves thus obtained, the intensity of the alpha-transition was determined to be $0.2 \pm 0.08\%$. The excitation energy of the second level of the Ra^{228} nucleus is 185 ± 5 kev.

Figure 7 shows the alpha spectrum of U^{238}. This spectrum was taken over a period of 30 hours. The alpha-transition to the second excited level is clearly evident. The intensity of the transition is $0.23 \pm 0.07\%$, and the excitation energy of the level is 160 ± 5 kev.

The ratios of the energies of the first and second levels for these isotopes are in good agreement with the theoretical values obtained on the basis of the collective model of the nucleus.[16] Hence there are good grounds for taking the first and second levels to be rotational, with characteristics 2^+ and 4^+ respectively.

3. COMPARISON OF EXPERIMENTAL VALUES WITH THEORY

In this work we used the formulas of Nosov, who kindly showed us his paper before publication.[17] Nosov's formulas are simple and convenient for the purposes of comparison with experiment. From the comparison of the theoretical values of transition probabilities to the 2^+ level with the experimental values, we determined the deformation parameters of the nuclei Ra^{228}, Th^{230}, and Th^{234} (see column 3 of the table). These values of deformation parameter make it possible to derive theoretical values for the transition probabilities to the 4^+ levels (column 4). Column 5 contains the experimental values of the transition probabilities to the 4^+ level. This table includes also our previous data[18] on Th^{230}.

From the table it can be seen that, within the limits of the errors of measurement, the experimental values of transition probability to the 4^+ level are satisfactorily related to the calculated values. Best agreement is obtained for $r_0 > 1.4$. This agrees with measurements of alpha-particle interactions with nuclei.[19]

CONCLUSION

The high energy resolution of gridded ionization chambers, together with their satisfactory stability, have made possible a study of the previously unobserved alpha transitions to the first and second excited levels of daughter nuclei in the alpha spectra of Th^{232} and U^{238}. It has been shown that these levels are rotational.

Decay schemes for Th^{232} and U^{238}, constructed from the results of this study, are shown in Fig. 8. The half lives for the fundamental transition have been taken from the literature.[20,21]

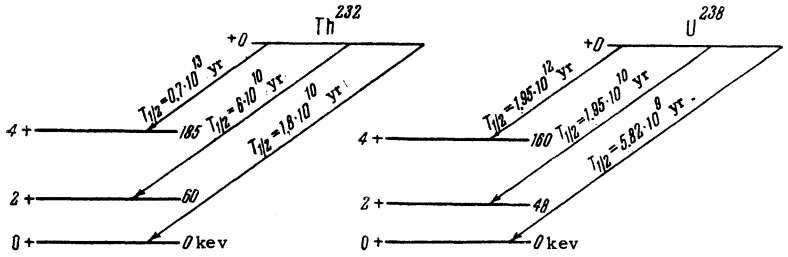

FIG. 8. Decay schemes for Th^{232} and U^{238}.

Daughter nucleus	Nuclear radius R_0 in units of $10^{-13} A^{1/3}$	Deformation Parameter	Transition probability to the 4^+ level (relative to the fundamental transition)	
			Theory	Experiment
Ra^{228}	1.0	0.27	0.14	
	1.4	0.21	0.17	0.26±0.1
	1.6	0.19	0.19	
Th^{230}	1.0	0.23	0.34	
	1.4	0.19	0.39	0.48±0.2
	1.6	0.18	0.41	
Th^{234}	1.0	0.21	0.15	
	1.4	0.17	0.21	0.3 ±0.09
	1.6	0.15	0.23	

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