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STUDIES OF α-SPECTRA IN ²²¹Fr, ²¹⁷At, ²¹³Bi AND ²¹³Po DECAYS

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1. Introduction

In the last ten years several investigations on the decays of nuclides from the 225 Ac equilibrium decay chain (Fig. 1) have been published. New data on the structure of the 217 At, 213 Bi, 213 Po and 209 Pb nuclei close to double magic 208 Pb have been gained. It is expected that the 225 Ac α -decay studies can reveal some new information on the presense of the static octupole deformation in the 221 Fr nucleus.

To study nuclear radiations of the above nuclei, ²²⁵Ac is separated from ²²⁹Th. The daughter nuclides are rapidly accumulated in the prepared source. Complex α -, β - and γ -radiation spectra and relatively short halflives of the daughter nuclei hinder the identification of specific transitions with the decay of appropriate nuclei from the ²²⁵Ac chain.

The fine structure lines in the ²²¹Fr α -decay were identified by Liang [1]. as he investigated the ²²⁵Ac α -recoil nuclei α -spectrum with the magnetic spectrograph. Ardisson et al. [2,3] developed and used fast radiochemical methods for separation of ²¹³Bi, ²⁰⁹Tl and ²²¹Fr nuclei and investigation of their γ -spectra. Sheline et al. [4] and Gromov et al. [5-8] confirmed the belonging of γ -transitions to the ²²⁵Ac, ²²¹Fr and ²¹⁷At decay in $(\alpha - \gamma)$ coincidence experiments.

But some problems still réquire caréful studies of the weak components of the 221 Fr, 217 At and 213 Bi α -spectra. For example,

Liang [1], when studying the α -spectrum of recoil nuclei from the ²²⁵Ac source, observed a weak line with $E_{\alpha} = 6037$ keV, $J_{\alpha} = 0.003$ % per decay. An excited ²¹⁷At level with energy 310 keV and $I^{\pi} = (13/2^+)$ is introduced on this basis [1,4,6]. Unlike the case with other levels introduced on the basis of the ²²¹Fr fine structure α -lines [1], no γ -transitions from the 310 keV level are observed. It is not impossible that the 6037 keV line is not associated with the ²²¹Fr α -decay, but with the α -decay of the daughter ²¹⁷At nucleus to the 1050 keV level of ²¹³Bi and, therefore, this line can be found in the α -spectrum of ²¹⁷At.

- Ardisson et al. [2] assumed that the 868 keV ²⁰⁹Tl level is excited in the ²¹³Bi α -decay. The sum intensity of the 868 and 545 keV γ - rays from this level was determined to be 0.03.% per decay. Accordingly, the fine structure α -line, with E_{α} =5018 keV and J_{α} =0.03 %, should be observed in the α -spectrum of ²¹³Bi.

- Chumin et al. [7], studying $(\alpha - \gamma)$ -coincidence in the decay of ²²⁵Ac







Fig. 2. Block diagram of the experiments for α -spectra investigation using an α -recoil: once (a) and twice (b).

S - ²²⁵Ac source, C1 and C2 - collectors of recoil nuclei, D - α -particle detector, RN - recoil nuclei



and its daughters, observed the coincidences of 150 keV γ -rays with $E_{\alpha 150}$ =6612 keV α -particles of ²²¹Ra, resulting from the β -decay of ²²¹Fr [1]. Thus, the observation of the ²²¹Fr β -decay [1] was confirmed. Its intensity was redetermined as (11±5)·10⁻⁵ decays. It is of interest to confirm this result in direct α -spectrum measurements.

- Chumin et al. [8] found the ²¹⁷At α -decay to the ²¹³Bi 759 keV level. The α -line $E_{\alpha759}$ (²¹⁷At)=6322 keV, $J_{\alpha759} = 5 \cdot 10^{-3}$ % is close to the ²²¹Fr α_0 -line ($E_{\alpha0} = 6341$ keV, $J_{\alpha0} = 85$ %) and was observed only in ($\alpha - \gamma$)-coincidences. It is worthwhile to confirm these data in direct α -spectrum measurements.

In the present paper the weak components of the 221 Fr, 217 At, and 213 Bi α -spectra are studied. The phenomenon of recoil in α -decay is used to eliminate the contribution to these spectra from the α -radiation of mother nuclei.

2. Experimental set-up

The main source of α -radiation was ²²⁵Ac separated from ²²⁹Th by the technique "The isotope generator of ²²⁵Ac" [9]. The ²²⁵Ac source activity was about 20 mCi. The ²²⁵Ac activity was electrolytically deposited on a tantalum foil and then vacuum evaporated on an aluminum foil. Small thickness of the resulting sources provided a considerable (up to 30 %) yield of recoil nuclei. To study α -spectra of the recoil nuclei, the ²²⁵Ac source was placed in a vacuum chamber so that the detector, situated in the chamber, could not detect α -particles from the source (Fig. 2). The recoil nuclei from the α -decay of ²²⁵Ac and daughter nuclei were gathered on a collector (C1 in Fig. 2). The detector recorded α -particles from the α -spectrum of the ²²¹Fr and daughter nuclei free of the contribution from α -particles of ²²⁵Ac was provided (from here on it is called the ²²¹Fr α -spectrum).

To have the ²¹⁷At α -spectrum the α -recoil phenomenon was used twice. The detector was placed in the chamber in a position where it could not "see" both the ²²⁵Ac source and the first collector. The recoil nuclei from the α -decay on the first collector were gathered on the second collector. The detector recorded α -particles from the decay of nuclei on the second collector (Fig. 2(b)).

A Canberra Si(Au) detector (diameter 10 mm, FWHM 15 keV) was used to measure α -spectra. The ²²¹Fr and ²¹⁷At decay spectra shown in Figs.

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3(b.c), are compared with the α -spectrum of the ²²⁵Ac and daughter nuclei (Fig. 3(a)). Note that the widths and forms of the α -lines from the decay of different nuclides are different. This is because part of the nuclei resulting from the α -decay penetrate into the collector material, which broadens the α -lines of these nuclei. That is why the narrowest in spectrum Fig. 3(a) lines belong to the ²²⁵Ac decay, in Fig. 3(b) to the ²²¹Fr decay, and in Fig. 3(c) to the ²¹⁷At decay. Note also that since the recoil nuclei leave the ²²⁵Ac source or the collectors with a relatively high yield, the number of decays recorded per time unit will be not constant for different members of the ^{225}Ac decay chain. It decreases with increasing number of α -decays leading to formation of the nucleus in question from ²²⁵Ac. Therefore, the relative intensities of α -lines from the decay of nuclei with the same mass numbers were used in the analysis of the result given in Tables 1 and 2. The relative intensities of α -lines were taken to be proportional to their areas, i.e. the efficiency of the detector was taken to be constant in the energy interval $E_{\alpha} = 5.0 \div 8.5$ MeV.

3. Experimental Results

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The comparison of the ²²¹Fr and ²¹⁷At spectra (Figs. 3(b,c)) with the spectrum of the ²²⁵Ac and daughter nuclei (Fig. 3(a)) allows one to estimate the degree of their purity from α -radiation of mother nuclei. In the ²¹⁷At α -spectrum the $E_{\alpha 0}$ =6341 keV ²²¹Fr line is observed. Its intensity is $5 \cdot 10^{-4} J_{\alpha_0} {}^{217}$ At. Thus the investigations, whose results are displaced in Fig. 3(c), are equivalent to the investigations with the mass-separated source of ²¹⁷At (32 ms) with the ²²¹Fr impurity of the order of $5 \cdot 10^{-4}$. To evaluate the ²²⁵Ac impurity is more difficult because the α -lines ($E_{\alpha 0}({}^{225}\text{Ac})=5830$ keV and $E_{\alpha 0}({}^{213}\text{Bi})=5870$ keV) are too close in energy. But it can be said with confidence that the intensity of the $E_{\alpha 0}({}^{225}\text{Ac})$ line in these spectra is below $5 \cdot 10^{-3}$.

Table 1 gives the results of the analysis of the spectra from Figs. 3(b) and (c).



Fig. 3. Alpha spectra of:

a) ²²⁵Ac and daughter nuclei (exposition 108 h);

b) ²²¹Fr and daughter nuclei (once recoil nuclei, exposition 270 h);

c) ²¹⁷At and daughter nuclei (twice recoil nuclei,exposition 182 h). Complex line: $E_0(^{221}\text{Fr}) = 6341$ keV and $E_{\alpha759}(^{217}\text{At}) = 6322$ keV is shown in the insert Table 1. Intensities of the α -lines of the ²¹⁷At, ²¹³Bi, and ²¹³Po decay

| Energy, keV | | Intensity per cent | | |
|---|---------------------|--------------------|--------------------|--|
| Levels | α -particles | Present paper | Other publications | |
| $^{217}\text{At} \rightarrow ^{213}\text{Bi}$ | | | | |
| 0 | 7067 | >99.9 | >99.9 | |
| 258 | 6814 | 0.038(4) | 0.036(3) [8] | |
| (465) | 6609 | - | 0.010(5) [10] | |
| 593 | 6485 | 0.022(2) | 0.021(2) [8] | |
| 759 | 6322 | 0.012(6) | 0.005(1) [8] | |
| (1050) | 6037 | < 0.002 | (0.003) $[1,10]$ | |
| $^{213}\text{Bi} \rightarrow ^{209}\text{Tl}$ | | | | |
| 0 | 5869 | 2.05(3) | 1.94(11) [10] | |
| 324 | 5549 . | 0.153(3) | 0.16(3) [10] | |
| (868) | (5018) | <10-4 | (0.03) $[2,10]$ | |
| $^{213}Po \rightarrow ^{209}Pb$ |) | | | |
| 0 | 8376 | 97.76(3) | 97.91(3) [10] | |
| 779 | 7614 | 0.0030(2) | 0.0047(5) [10] | |

Note: Energies of alpha-particles and levels are from references [1,2,8,10].

- The ²¹⁷At α -spectrum: Intensities of the $E_{\alpha 258} = 6814$ keV and $E_{\alpha 593}=6485$ keV lines are in good agreement with the results of the $(\alpha - \gamma)$ -coincidence experiments [8]. The $E_{\alpha} = 6341$ keV line (Fig. 3(c)), whose main part we attribute to the ²²¹Fr decay (see above), is a complex one. Its decomposition, shown in the insert in Fig. 3(c), allowed us to determine the intensity of the new $E_{\alpha 759} = 6322$ keV line (α -decay of ²¹⁷At in the 759 keV level of ²¹³Bi). It is $J_{\alpha 759} = (12\pm6)\cdot10^{-3}$ % and agrees with [8]. The intensity evaluation of the $E_{\alpha} = 6037$ keV line does not exclude the possibility of assigning this α -line to the ²¹⁷At decay.

- The ²¹³Bi α -spectrum: The measured intensities of the $E_{\alpha 0}$ (²¹³Bi)= 5870 keV and $E_{\alpha 324}$ (²¹³Bi)=5549 keV lines agree with the known ones [10]. The measured upper limit for the intensity of the $E_{\alpha 868}$ =5018 keV line appeared to be 100 times smaller than expected from [2]. Thus, the assumption that the ²⁰⁹Tl 868 keV level is excited in the ²¹³Bi α -decay is not confirmed. Note that in their later paper [3] the authors of [2] attributed the 868 keV γ -transition to the ²¹³Bi \rightarrow ²⁰⁹Po β -decay, but did not abandon the earlier assumption that the 868 keV

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level is excited in 209 Tl. Accordingly, in the 1996 Table of Isotopes [10] the 868 keV level in 209 Tl is preserved.

- The ²¹³Po α -spectrum: The measured intensity of the $E_{\alpha 778}(^{213}\text{Po})$ line agrees with the known data [10].

Table 2 gives the results concerning the $\beta^-{\rm -decay}$ of $^{221}{\rm Fr},~^{217}{\rm At}$ and $^{213}{\rm Bi}.$

| Table | 2. | Intensities of | f the | 221 Fr, | ²¹⁷ At. | , and ²¹³ | ³ Bi | β^{-} - | deca | ١ |
|-------|-----------|----------------|-------|--------------|--------------------|----------------------|-----------------|---------------|------|---|
|-------|-----------|----------------|-------|--------------|--------------------|----------------------|-----------------|---------------|------|---|

| Nuclei | β -decay | | | | |
|-------------------|-------------------------------|------------------------------|--|--|--|
| | Present paper | Other publications | | | |
| ²²¹ Fr | $(4.8 \pm 1.5) \cdot 10^{-5}$ | $(11\pm5)\cdot10^{-5}$ [7] | | | |
| ²¹⁷ At | $(6.7\pm2.4)\cdot10^{-5}$ | $(12\pm 6)\cdot 10^{-5}$ [1] | | | |
| | | $<5.10^{-5}$ [7] | | | |
| ²¹³ Bi | (0.9776(3) | 0.9791(3)[10] | | | |

- ²²¹**Fr**: In the ²²¹Fr α -spectrum very weak lines of ²²¹Ra are observed. The $E_{\alpha 150}$ = 6612 keV line is the most distinct. This line was earlier attributed in the ²²⁵Ac decay chain to the ²¹⁷At decay [10]. Its belonging to the ²²¹Ra decay was proved in [7] by observation of coincidences of this line with the 150 keV γ -ray, and is confirmed in the present study by the fact that ²²¹Ra α -lines are displayed in the ²²¹Fr spectrum and are not observed in the ²¹⁷At spectrum. In the ²²⁵Ac decay chain ²²¹Ra results from the β -decay of ²²¹Fr. There is not another explanation of the ²²¹Ra presence in the ²²⁵Ac decay chain. Using the 6612 keV α -line intensity we determined the intensity of the ²²¹Fr β -decay branch to be (4.8± 1.5)·10⁻³ % in agreement with the result of [7]: (11±5)·10⁻³ %. Now that the 6612 keV α -line is ascribed to the ²²¹Ra α -decay, there are not experimental data for introduction of the ²¹³Bi 450 keV level in the decay of ²¹⁷At [10].

- ²¹⁷At: In the ²¹⁷At α -spectrum, (Fig. 3(c)) the $E_{\alpha 0}$ =7741 keV, $J_{\alpha 0}$ =100 % α_0 -line of ²¹⁷Rn ($T_{1/2}$ =0.54 ms) is observed. This ²¹⁷Ra is formed both in the ²²¹Ra α -decay and in the β -decay of ²¹⁷At. The intensity of the ²¹⁷At β -decay branch was calculated as a difference between the $E_{\alpha 0}$ =7741 keV line intensity in the spectrum of Fig. 3(c) and the ²²¹Fr β -decay branch intensity found above. The value $(6.7\pm2.4)\cdot10^{-5}$ per decay does not contradict the upper limit determined in [7] and earlier investigations.

- ²¹³Bi: The intensity of the ²¹³Bi α-decay determined from the ratio of the α-line areas in the spectrum of Fig. 3(c):

$$\frac{S_{\alpha 0}(^{213}Po)}{S_{\alpha 0}(^{213}Po) + S_{\alpha 0}(^{213}Bi) + S_{\alpha 324}(^{213}Bi)} = 0.9776(10)$$

is in agreement with the known value [10].

4. Conclusion

The use of the α -recoil phenomenon to study α -spectra in the ²²⁵Ac equilibrium decay chain has allowed us to free spectra investigated from the α -radiation of mother nuclei and to gain new or more reliable experimental data on the intensity of weak components of these spectra.

The research of the ²¹⁷At α -spectrum confirms the results [8] about the excitation of the 759 keV level in ²¹³Bi and yields more correct data on the intensity of the ²¹⁷At α -decay to the 258 and 593 keV levels in ²¹³Bi. It is established that the 6612 keV α -line, previously attributed to the ²¹⁷At α -decay, arises from the ²²¹Ra α -decay and thus there is no experimental basis for the introduction of the 450 keV level in ²¹³Bi.

It is shown that the assumed [2] excitation of the 209 Tl 868 keV level in the 213 Bi α -decay contradicts the results of the present 213 Bi α -spectrum investigation.

Identification of the ²²¹Ra and ²¹⁷Rn α -lines in α - spectra of the nuclei from the ²²⁵Ac decay chain and measurement of their intensity has allowed us to repeat determination of the β -decay intensity for ²²¹Fr, ²¹⁷At, and ²¹³Bi.

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Чумин В.Г. и др. Исследование α -спектров при распаде ²²¹ Fr, ²¹⁷ At, ²¹³ Bi и ²¹³ Po

Для получения информации об интенсивности слабых компонентов α -спектров нуклидов из цепочки распадов ²²⁵Ac использовано явление отдачи. при α -распаде. Показано, что нет экспериментальных оснований для введения уровней 450 кэВ ²¹³Bi при распаде ²¹⁷At-и 868 кэВ ²⁰⁹Tl при распаде. ²¹³Bi. Подтверждается возбуждение уровня 759 кэВ ²¹³Bi при распаде ²¹⁷At. Измерены интенсивности β^{-} -распада ²²¹Fr, ²¹⁷At-и ²¹³Bi.

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Studies of α -Spectra in ²²¹Fr, ²¹⁷At, ²¹³Bi and ²¹³Po Decays

The alpha-recoil phenomenon is used to gain data on the weak components of the α -spectra of the nuclides from the ²²⁵Ac equilibrium chain. It is established that there is no experimental basis for introducing the 450 keV level of ²¹³Bi in the ²¹⁷At decay and the 868 keV level of ²⁰⁹Tl in the ²¹³Bi decay. Excitation of the 759 keV level in the ²¹⁷At decay is confirmed. The intensities of the ²²¹Fr, ²¹⁷At and ²¹³Bi β -decay are measured.

The investigation has been performed at the Laboratory of Nuclear Problems, JINR.

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