

ОБЪЕДИНЕННЫЙ ИНСТИТУТ ЯДЕРНЫХ ИССЛЕДОВАНИЙ

Дубна

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E6-97-189

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STUDIES OF  $\alpha$ -SPECTRA IN  $^{221}$ Fr,  $^{217}$ At,  $^{213}$ Bi AND  $^{213}$ Po DECAYS

Submitted to «Известия РАН, серия физическая»



Swy749

## 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  $\alpha$ -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,  $^{225}$ Ac is separated from  $^{229}$ Th. The daughter nuclides are rapidly accumulated in the prepared source. Complex  $\alpha$ -,  $\beta$ - and  $\gamma$ -radiation spectra and relatively short half-lives of the daughter nuclei hinder the identification of specific transitions with the decay of appropriate nuclei from the  $^{225}$ Ac chain.

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

But some problems still require careful studies of the weak components of the  $^{22t}$ Fr,  $^{217}$ At and  $^{213}$ Bi  $\alpha$ -spectra. For example,

- Liang [1], when studying the  $\alpha$ -spectrum of recoil nuclei from the  $^{225}$ Ac source, observed a weak line with  $E_{\alpha}=6037~{\rm keV},\ J_{\alpha}=0.003$ % per decay. An excited  $^{217}$ At 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  $^{221}$ Fr fine structure  $\alpha$ -lines [1], no  $\gamma$ -transitions from the 310 keV level are observed. It is not impossible that the 6037 keV line is not associated with the  $^{221}$ Fr  $\alpha$ -decay, but with the  $\alpha$ -decay of the daughter  $^{217}$ At nucleus to the 1050 keV level of  $^{213}$ Bi and, therefore, this line can be found in the  $\alpha$ -spectrum of  $^{217}$ At.
- Ardisson et al. [2] assumed that the 868 keV <sup>209</sup>Tl level is excited in the <sup>213</sup>Bi  $\alpha$ -decay. The sum intensity of the 868 and 545 keV  $\gamma$  rays from this level was determined to be 0.03 % per decay. Accordingly, the fine structure  $\alpha$ -line, with  $E_{\alpha}$ =5018 keV and  $J_{\alpha}$ =0.03 %, should be observed in the  $\alpha$ -spectrum of <sup>213</sup>Bi.
- Chumin et al. [7], studying  $(\alpha \gamma)$ -coincidence in the decay of  $^{225}{\rm Ac}$

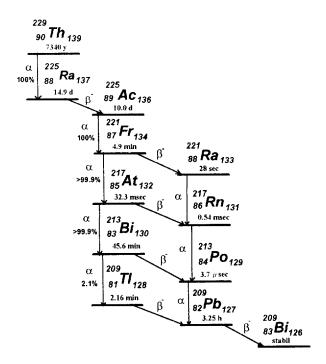


Fig. 1. Decay chain of <sup>229</sup>Th to <sup>209</sup>Bi

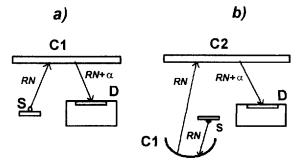


Fig. 2. Block diagram of the experiments for  $\alpha$ -spectra investigation using an  $\alpha$ -recoil: once (a) and twice (b).

S -  $^{225}{\rm Ac}$  source, C1  $\,$  and C2 - collectors of recoil nuclei, D -  $\alpha$  -particle detector, RN - recoil nuclei

and its daughters, observed the coincidences of 150 keV  $\gamma$ -rays with  $E_{\alpha 150}$ =6612 keV  $\alpha$ -particles of  $^{221}$ Ra, resulting from the  $\beta$ -decay of  $^{221}$ Fr [1]. Thus, the observation of the  $^{221}$ Fr  $\beta$ -decay [1] was confirmed. Its intensity was redetermined as  $(11\pm5)\cdot10^{-5}$  decays. It is of interest to confirm this result in direct  $\alpha$ -spectrum measurements.

- Chumin et al. [8] found the <sup>217</sup>At  $\alpha$ -decay to the <sup>213</sup>Bi 759 keV level. The  $\alpha$ -line  $E_{\alpha 759}$  (<sup>217</sup>At)=6322 keV,  $J_{\alpha 759}=5\cdot 10^{-3}$  % is close to the <sup>221</sup>Fr  $\alpha_0$ -line ( $E_{\alpha 0}=6341$  keV,  $J_{\alpha 0}=85$  %) and was observed only in ( $\alpha - \gamma$ )-coincidences. It is worthwhile to confirm these data in direct  $\alpha$ -spectrum measurements.

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

## 2. Experimental set-up

The main source of  $\alpha$ -radiation was  $^{225}\mathrm{Ac}$  separated from  $^{229}\mathrm{Th}$  by the technique "The isotope generator of  $^{225}\mathrm{Ac}$ " [9]. The  $^{225}\mathrm{Ac}$  source activity was about 20 mCi. The  $^{225}\mathrm{Ac}$  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  $\alpha$ -spectra of the recoil nuclei, the  $^{225}\mathrm{Ac}$  source was placed in a vacuum chamber so that the detector, situated in the chamber, could not detect  $\alpha$ -particles from the source (Fig. 2). The recoil nuclei from the  $\alpha$ -decay of  $^{225}\mathrm{Ac}$  and daughter nuclei were gathered on a collector (C1 in Fig. 2). The detector recorded  $\alpha$ -particles from the decay of the recoil nuclei gathered on the collector. Thus the  $\alpha$ -spectrum of the  $^{221}\mathrm{Fr}$  and daughter nuclei free of the contribution from  $\alpha$ -particles of  $^{225}\mathrm{Ac}$  was provided (from here on it is called the  $^{221}\mathrm{Fr}$   $\alpha$ -spectrum).

To have the  $^{217}$ At  $\alpha$ -spectrum the  $\alpha$ -recoil phenomenon was used twice. The detector was placed in the chamber in a position where it could not "see" both the  $^{225}$ Ac source and the first collector. The recoil nuclei from the  $\alpha$ -decay on the first collector were gathered on the second collector. The detector recorded  $\alpha$ -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  $\alpha$ -spectra. The  $^{221}\mathrm{Fr}$  and  $^{217}\mathrm{At}$  decay spectra shown in Figs.

3(b,c), are compared with the  $\alpha$ -spectrum of the  $^{225}\mathrm{Ac}$  and daughter nuclei (Fig. 3(a)). Note that the widths and forms of the  $\alpha$ -lines from the decay of different nuclides are different. This is because part of the nuclei resulting from the lpha-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 <sup>225</sup>Ac decay, in Fig. 3(b) to the <sup>221</sup>Fr decay, and in Fig. 3(e) to the  $^{247}\mathrm{At}$  decay. Note also that since the recoil nuclei leave the  $^{225}\mathrm{Ac}$  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  $^{225}\mathrm{Ac}$  decay chain. It decreases with increasing number of  $\alpha$ -decays leading to formation of the nucleus in question from <sup>225</sup>Ac. Therefore, the relative intensities of  $\alpha$ -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  $\alpha$ -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  $E_{\alpha} = 5.0 \pm 8.5 \text{ MeV}.$ 

# 3. Experimental Results

The comparison of the  $^{221}$ Fr and  $^{247}$ At spectra (Figs. 3(b,c)) with the spectrum of the  $^{225}$ Ac and daughter nuclei (Fig. 3(a)) allows one to estimate the degree of their purity from  $\alpha$ -radiation of mother nuclei. In the  $^{247}$ At  $\alpha$ -spectrum the  $E_{\alpha0}$ =6341 keV  $^{221}$ Fr line is observed. Its intensity is  $5 \cdot 10^{-4}$  J<sub> $\alpha$ </sub>,  $^{247}$ At. Thus the investigations, whose results are displacd in Fig. 3(c), are equivalent to the investigations with the mass-separated source of  $^{247}$ At (32 ms) with the  $^{221}$ Fr impurity of the order of  $5 \cdot 10^{-4}$ . To evaluate the  $^{225}$ Ac impurity is more difficult because the  $\alpha$ -lines ( $E_{\alpha0}(^{225}$ Ac)=5830 keV and  $E_{\alpha0}(^{213}$ Bi) =5870 keV) are too close in energy. But it can be said with confidence that the intensity of the  $E_{\alpha0}(^{225}$ 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).

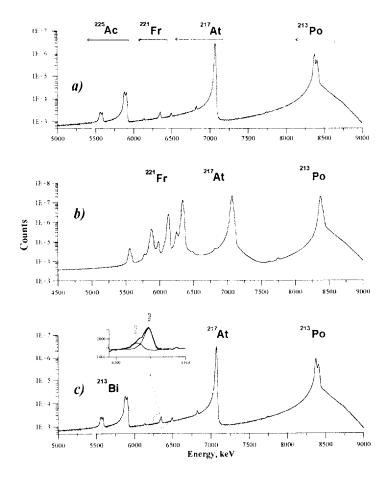


Fig. 3. Alpha spectra of:

- a)  $^{225}\mathrm{Ac}$  and daughter nuclei (exposition 108 h);
- b) <sup>221</sup>Fr and daughter nuclei (once recoil nuclei, exposition 270 h);
- c)  $^{217}\mathrm{At}$  and daughter nuclei (twice recoil nuclei, exposition 182 h).

Complex line:  $E_0(^{224}{\rm Fr})=6341~{\rm keV}$  and  $E_{\alpha759}(^{217}{\rm At})=6322~{\rm keV}$  is shown in the insert

Table 1. Intensities of the  $\alpha$ -lines of the  $^{217}\mathrm{At},\,^{213}\mathrm{Bi},\,$  and  $^{213}\mathrm{Po}$  decay

| Energy, keV                                      |                     | Intensity per cent |                    |
|--|---------------------|--------------------|--------------------|
| Levels   | $\alpha$ -particles | Present paper      | Other publications |
| $^{217}{\rm At} - ^{213}{\rm B}$                 | i                   |                    |                    |
| 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]     |
| $^{213}\mathrm{Bi} \rightarrow ^{209}\mathrm{T}$ | 1                   |                    |                    |
| 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]      |
| <sup>213</sup> Po → <sup>209</sup> P             | 'b                  |                    |                    |
| ()   | 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  $^{217}$ At  $\alpha$ -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  $^{221}$ Fr 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 ( $\alpha$ -decay of  $^{217}$ At in the 759 keV level of  $^{213}$ Bi). 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  $\alpha$ -line to the  $^{217}$ At decay.
- The <sup>213</sup>Bi α-spectrum: The measured intensities of the E<sub>α0</sub> (<sup>213</sup>Bi)= 5870 keV and E<sub>α324</sub> (<sup>213</sup>Bi)=5549 keV lines agree with the known ones [10]. The measured upper limit for the intensity of the E<sub>α868</sub>=5018 keV line appeared to be 100 times smaller than expected from [2]. Thus, the assumption that the <sup>209</sup>Tl 868 keV level is excited in the <sup>213</sup>Bi α-decay is not confirmed. Note that in their later paper [3] the authors of [2] attributed the 868 keV γ-transition to the <sup>213</sup>Bi →<sup>209</sup>Po β-decay, but did not abandon the earlier assumption that the 868 keV

- level is excited in  $^{209}$ Tl. Accordingly, in the 1996 Table of Isotopes [10] the 868 keV level in  $^{209}$ Tl is preserved.
- The <sup>213</sup>Po  $\alpha$ -spectrum: The measured intensity of the  $E_{\alpha778}(^{213}\text{Po})$  line agrees with the known data [10].

Table 2 gives the results concerning the  $\beta^+$ -decay of  $^{224}{\rm Fr.}$   $^{217}{\rm At}$  and  $^{213}{\rm Bi}$ .

**Table 2.** Intensities of the  $^{221}$ Fr,  $^{217}$ At, and  $^{213}$ Bi  $\beta^-$ -decay

| Nuclei              | $\beta$ -decay            |                            |  |
|---------------------|---------------------------|----------------------------|--|
|                     | Present paper             | Other publications         |  |
| $^{221}\mathrm{Fr}$ | $(4.8\pm1.5)\cdot10^{-5}$ | $(11\pm5)\cdot10^{-5}$ [7] |  |
| $^{217}\mathrm{At}$ | $(6.7\pm2.4)\cdot10^{-5}$ | $(12\pm6)\cdot10^{-5}$ [1] |  |
|                     |                           | $<5.10^{-5}$ [7]           |  |
| <sup>213</sup> Bi   | (0.9776(3)                | 0.9791(3)[10]              |  |

- $^{221}$ Fr: In the  $^{221}$ Fr α-spectrum very weak lines of  $^{221}$ Ra are observed. The  $E_{\alpha 150}=6612$  keV line is the most distinct. This line was earlier attributed in the  $^{225}$ Ac decay chain to the  $^{217}$ At decay [10]. Its belonging to the  $^{221}$ Ra decay was proved in [7] by observation of coincidences of this line with the 150 keV  $\gamma$ -ray, and is confirmed in the present study by the fact that  $^{221}$ Ra α-lines are displayed in the  $^{221}$ Fr spectrum and are not observed in the  $^{217}$ At spectrum. In the  $^{225}$ Ac decay chain  $^{221}$ Ra results from the β-decay of  $^{221}$ Fr. There is not another explanation of the  $^{221}$ Ra presence in the  $^{225}$ Ac decay chain. Using the 6612 keV α-line intensity we determined the intensity of the  $^{221}$ Fr β-decay branch to be  $(4.8\pm 1.5)\cdot 10^{-3}$ % in agreement with the result of [7]:  $(11\pm 5)\cdot 10^{-3}$ %. Now that the 6612 keV α-line is ascribed to the  $^{221}$ Ra α-decay, there are not experimental data for introduction of the  $^{213}$ Bi 450 keV level in the decay of  $^{217}$ At [10].
- <sup>217</sup>**At:** In the <sup>217</sup>At α-spectrum, (Fig. 3(c)) the  $E_{\alpha 0}$ =7741 keV.  $J_{\alpha 0}$ =100 % α<sub>0</sub>-line of <sup>217</sup>Rn ( $T_{1/2}$ =0.54 ms) is observed. This <sup>217</sup>Ra is formed both in the <sup>221</sup>Ra α-decay and in the β-decay of <sup>217</sup>At. The intensity of the <sup>217</sup>At 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 <sup>221</sup>Fr 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.

 - <sup>213</sup>Bi: The intensity of the <sup>213</sup>Bi 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  $\alpha$ -recoil phenomenon to study  $\alpha$ -spectra in the  $^{225}{\rm Ac}$  equilibrium decay chain has allowed us to free spectra investigated from the  $\alpha$ -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  $^{217}$ At  $\alpha$ -spectrum confirms the results [8] about the excitation of the 759 keV level in  $^{213}$ Bi and yields more correct data on the intensity of the  $^{217}$ At  $\alpha$ -decay to the 258 and 593 keV levels in  $^{213}$ Bi. It is established that the 6612 keV  $\alpha$ -line, previously attributed to the  $^{217}$ At  $\alpha$ -decay, arises from the  $^{221}$ Ra  $\alpha$ -decay and thus there is no experimental basis for the introduction of the 450 keV level in  $^{213}$ Bi.

It is shown that the assumed [2] excitation of the  $^{209}$ Tl 868 keV level in the  $^{213}$ Bi  $\alpha$ -decay contradicts the results of the present  $^{213}$ Bi  $\alpha$ -spectrum investigation.

Identification of the  $^{221}\mathrm{Ra}$  and  $^{217}\mathrm{Rn}$   $\alpha$ -lines in  $\alpha$ - spectra of the nuclei from the  $^{225}\mathrm{Ac}$  decay chain and measurement of their intensity has allowed us to repeat determination of the  $\beta$ -decay intensity for  $^{221}\mathrm{Fr}$ ,  $^{247}\mathrm{At}$ , and  $^{213}\mathrm{Bi}$ .

The authors are grateful to Dr. V.B. Brudanin for his interest in this investigation. The investigation was supported by the Russian Foundation for Basic Research (Project No. 94-02-04828).

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Received by Publishing Department on June 9, 1997.

Чумин В.Г. и др. Исследование  $\alpha$ -спектров при распаде  $^{221}$ Fr,  $^{217}$ At,  $^{213}$ Bi и  $^{213}$ Po

Для получения информации об интенсивности слабых компонентов  $\alpha$ -спектров нуклидов из цепочки распадов <sup>225</sup>Ас использовано явление отдачи при  $\alpha$ -распаде. Показано, что нет экспериментальных оснований для введения уровней 450 кэВ <sup>213</sup>Ві при распаде <sup>217</sup>Аt и 868 кэВ <sup>209</sup>ТІ при распаде <sup>213</sup>Ві. Подтверждается возбуждение уровня 759 кэВ <sup>213</sup>Ві при распаде <sup>217</sup>Аt. Измерены интенсивности  $\beta$ --распада <sup>221</sup>Fr, <sup>217</sup>At и <sup>213</sup>Ві.

Работа выполнена в Лаборатории ядерных проблем ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна, 1997

Chumin V.G. et al. E6-97-189

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

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

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

Preprint of the Joint Institute for Nuclear Research. Dubna, 1997

Редактор Е.И.Кравченко. Макет Т.Е.Попеко

Подписано в нечать 02.07.97 Формат 60 × 90/16. Офсетная печать. Уч.-изд.листов 1,14 Тираж 335. Заказ 50034. Цена 1368 р.

Издательский отдел Объединенного института ядерных исследований Дубна Московской области