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ANALYSIS OF PARASITIC SIGNALS IN THE METHOD OF RECOIL NUCLEI APPLIED TO DIRECT OBSERVATION OF THE ^{229m}Th ISOMERIC STATE

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We carry out necessary theoretical justifications for the method of recoil nuclei in application to the direct observation of the ^{229m}Th isomeric state. We consider Cherenkov radiation, phosphorescence, and fluorescence in a crystal plate, which is used for collecting the thorium recoil nuclei, and discuss the ways to avoid these parasitic signals in order to successfully reveal ^{229m}Th decay photons.

Keywords: Th-229, recoil nuclei, isomeric state, γ-decay, Cherenkov radiation, phosphorescence, fluorescence, magnesium fluoride.

1. Introduction

The isotope ^{229}Th is of significant interest due to the predicted isomeric state with the energy astonishingly low for nuclear physics [1]. It is considered to lie in a range of several eV [2–4] typical of the transitions in atomic electron shells, so this feature could allow to explore correlations between atomic and nuclear degrees of freedom. According to the latest indirect investigations, this isomeric state has an energy of 7.8 ± 0.5 eV and a half-life of $\sim$5 h [4]. Till now, many fundamental theoretical investigations of the processes in atoms ^{229}Th are made [5–8]. In addition, such amazing applications as a novel frequency standard with relative accuracy 10^{-19} [9] and the first nuclear laser [10] are suggested even in spite of the fact that there are no reliable proofs that the ^{229m}Th isomeric state really exists.

Actually, several attempts to find the state ^{229m}Th were made, but with no success. Recently, Zhao *et al.* reported the first observation of the ^{229m}Th decay photons [11], but the result was called in question at once [12]. By now, the experiment has no unambiguous conclusions. In our opinion, the problem grows from the lack of theoretical justifications of the approach used therein. In this paper, we fill this gap, by making the necessary theoretical analysis.

The method used in Ref. [11] consists in implanting ^{229}Th nuclei recoiled from an α-decaying ^{233}U ($T_{1/2} = 1.592 \times 10^5$ yr) into a plate prepared from the material transparent in VUV (CaF_2, MgF_2, fused silica, *etc.*). The isotope ^{233}U decays with the $\sim$2% branching to the state ^{229m}Th [13]. The UV photons, which are emitted as a result of the isomeric state decay, are searched with a photomultiplier (PMT). In the experiment (see Ref. [11]), a uranium sample with an effective activity (that really takes part in implanting) of $\sim$170 kBq giving, by authors' estimations, $\sim$1000 isomers/s in the crystal plate was used. For the few-hour implantation period, this led to a signal of the order of 1 photon per second during the measurement with a PMT.

A PMT fixes not only the ^{229m}Th radiation. First of all, the phosphorescence in the crystal caused by α-radiation in the implanting process can take place. In addition, it was pointed out in Ref. [12] that if a uranium sample is not fresh enough, the daughter isotopes of ^{233}U are accumulated, by implanting their recoil nuclei into the plate. In turn, these recoil nu-

clei decay, by giving α- and β-particles leading to Cherenkov radiation and luminescence in the crystal. To make the method of recoil nuclei really consistent, all these contributions must be analyzed.

2. Cherenkov Radiation

If the uranium sample is not fresh enough, it has not only ^{233}U but its daughter nuclei as well, which give their recoil nuclei into the crystal plate. This may cause Cherenkov radiation. Actually, the threshold of this process in a transparent medium with the refractive index $n = 1.5$ is equal to 170 keV for radiating electron's kinetic energy. There are the fast-decaying nuclei of ^{213}Bi ($T_{1/2} = 45.59$ min) and ^{209}Pb ($T_{1/2} = 3.25$ h) in the decay chain of ^{233}U. They are β-radioactive with mean β-electron's energies 435 and 644 keV, respectively. So, Cherenkov radiation may take place.

The ^{233}U decay chain is a part of the neptunium series shown in Fig. 1. The half-lives of the isotopes, the energies of their decays, and the fractions of the decay branches are available. If the β-decay takes place, then the energy averaged over the spectrum of β-electrons can be written. The β-radioactive nuclei with the energies exceeding the Cherenkov threshold are presented in Table 1.

The other β-emitters either have a too small decay energy or do not belong to the main decay channel.

We consider the age of the uranium sample to be much smaller than the half-life of ^{229}Th ($T_{\mathrm{Th}} = 7400$ yr) and much greater than the half-lives of ^{217}At, ^{213}Po, and the other nuclei after ^{229}Th. We note also that ^{229}Th decays much faster than ^{233}U. Under these conditions, the isotopic composition of the sample at the moment of the experiment may be considered as established and defined by the number of ^{229}Th nuclei, which is estimated as $N_{\mathrm{Th}}(t) = \lambda_{\mathrm{U}} N_{\mathrm{U}} t$, where $\lambda_{\mathrm{U}} = \frac{\ln 2}{T_{\mathrm{U}}}$ is the rate of decay with the half-life T_{U}, N_{U} denotes the number of uranium nuclei, and t is the time of storing of the sample. The activity of thorium is given by

$$A_{\mathrm{Th}}(t) = \lambda_{\mathrm{Th}} N_{\mathrm{Th}}(t) = \frac{\ln 2}{T_{\mathrm{Th}}} A_{\mathrm{U}} t. \qquad (1)$$

The process of collection of the recoil nuclei in the crystal plate does not change the total isotopic equilibrium (in the crystal plate and out of it). Let us assume that this process is carried out until the number of the isotopes in the plate becomes established.

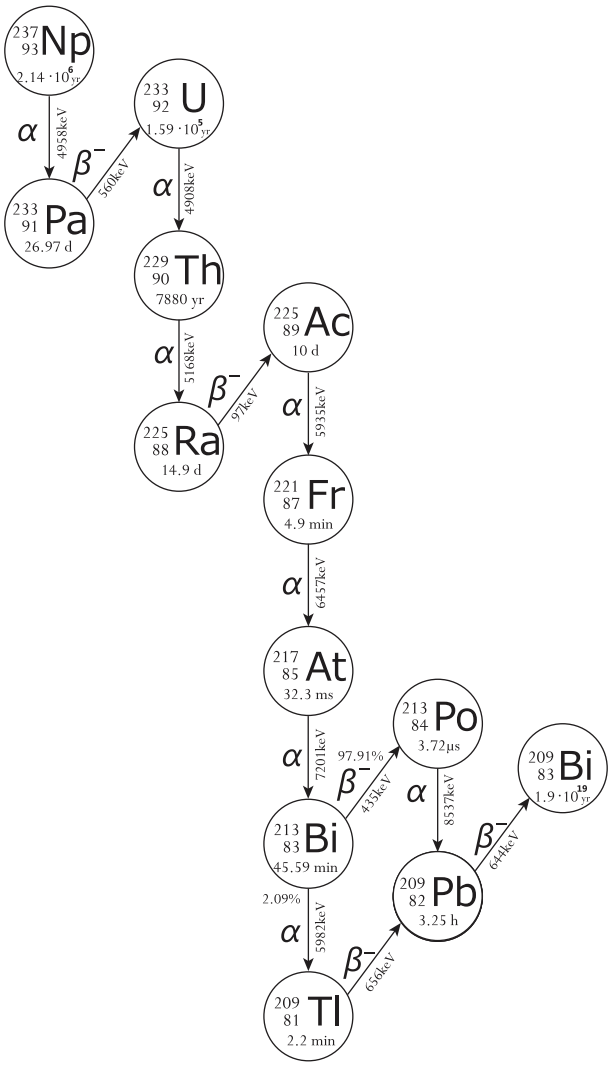

Fig. 1. Neptunium series

Table 1. **β-active isotopes**

Isotope	Half-life	E_e (average)	Rate of the channel
^{213}Bi	45.59 min	435 keV	97,8%
^{209}Pb	3.25 h	644 keV	100%

We consider that ^{213}Bi and ^{209}Pb in the plate constitute a half of their total number, because, approximately, the half has recoil impulses directed to the plate. Using this fact and the equilibrium condition (activities of the isotopes are equal), we deduce the flux of β-electrons in the plate from each isotope caus-

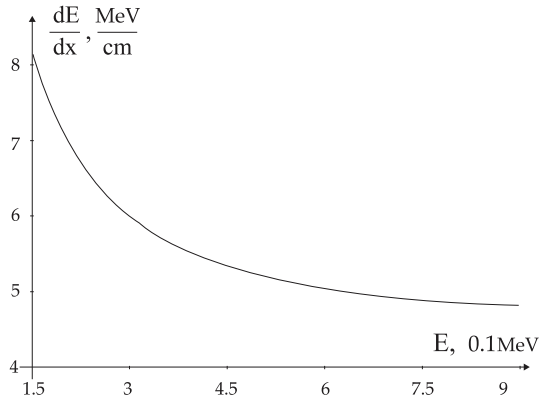

Fig. 2. Stopping force for an electron in MgF_2

ing Cherenkov radiation in the following form:

$$A_{\rm Bi} = A_{\rm Pb} = \frac{1}{2}A_{\rm Th}(t) = \frac{1}{2}\frac{\ln 2}{T_{\rm Th}}A_{\rm U}t. \quad (2)$$

In Ref. [11], the sample with the activity $A_{\rm U} = 170$ kBq was observed during ∼100 days. From (2), we calculate that the nuclei, which cause Cherenkov radiation, have the activity $A_{\rm Bi} = A_{\rm Pb} = A \approx 2.2$ Bq.

The Cherenkov loss of electron's energy per 1 cm is given by the formula [14]

$$\delta E = \frac{e^2}{c^2}\int\limits_{\beta n(\omega)>1} \omega\Big[1 - \frac{1}{\beta^2 n^2(\omega)}\Big]d\omega, \quad (3)$$

where e is the elementary charge, c is the speed of light, β is electron's speed in units of c, the integration is carried out over the frequency ω, and $n(\omega)$ denotes the spectral dependence of the refractive index. For $n(\omega) = n$, we immediately obtain the number of irradiated photons per 1 cm in the range of photons' energies of ΔE:

$$\delta N = \frac{e^2}{\hbar^2 c^2}\Big[1 - \frac{1}{\beta^2 n^2}\Big]\Delta E. \quad (4)$$

Using the electronic database [15], we obtained the loss of electron's energy (ionization + radiation) in the crystal MgF_2 as a function of the energy (Fig. 2). The total number of Cherenkov photons produced by an electron in the process of stopping from the initial energy to the energy of the barrier $E_{\rm min} = 170$ keV equals

$$N = \int \frac{dN}{dE}dE = \int \frac{dN/dx}{dE/dx}dE, \quad (5)$$

where the integration is carried out over the range of energies from the initial E_e to the Cherenkov barrier $E_{\rm min}$. Taking (4) into account, we obtain

$$N = \frac{\alpha\Delta E}{\hbar c}I, \quad (6)$$

$$I = \int \frac{1}{dE/dx}\Big[1 - \frac{1}{n^2(1 - \frac{1}{(1+E/m_e c^2)})}\Big]dE. \quad (7)$$

Here, $\alpha = \frac{e^2}{\hbar c} = 1/137$ is the fine-structure constant, and ΔE is taken to be the pass band of a Hamamatsu R8486 PMT used in Ref. [11], which is equal to 6 eV. The results of numerical calculations are given in Table 2.

Table 2. **Total number of photons per 1 electron**

β-source	I, 10^{-3} cm	N
^{213}Bi	10.9	24
^{209}Pb	27.8	62

The total intensity of Cherenkov radiation is the sum over the β-electrons produced in the plate per second:

$$A_{\rm total} = \frac{1}{2}(A_{\rm Bi}N_{\rm Bi} + A_{\rm Pb}N_{\rm Pb}) = \frac{A}{2}(N_{\rm Bi} + N_{\rm Pb}). \quad (8)$$

The factor 1/2 appears, because the recoil nuclei stick near the surface of the crystal, and a half of β-electrons fly out from the crystal at once after the production. Using (8), we obtain $A_{\rm total} \approx 95$ photons/s. In Ref. [11], the 1% – optical coupling to a PMT sensor and the 10% – PMT quantum efficiency were assumed. Under these assumptions, a PMT must register ∼0.1 Cherenkov photons per second. It is much smaller than the expected useful signal in Ref. [11].

This rough estimation shows that one may neglect Cherenkov radiation caused by the β-decaying daughter nuclei of ^{233}U, if the sample was purified at the earliest 100 days ago. There is another way to understand whether Cherenkov radiation gives a significant contribution or not. From (4), we find that Cherenkov photons are distributed uniformly over the spectrum. This fact allows us to verify whether this parasitic signal takes place or not. Actually, the following rule holds: *if the investigated range of energies has a subrange with no PMT signal, then there is no Cherenkov*

radiation at all. It is especially useful, if there are not only the daughter nuclei of ^{233}U, but some other sources of Cherenkov radiation.

The experiment (see Ref. [11]) was carried out with MgF_2 plates with implanted ^{229}Th, being explored with two PMTs: a Hamamatsu R8487 PMT (operating range 115–195 nm) and a Hamamatsu R8486 one (operating range 115–320 nm). The first PMT showed no signal. So, in accordance with the rule, we conclude that there was no Cherenkov radiation in contradiction with the comment in Ref. [12].

3. Phosphorescence

Even if the uranium sample is quietly fresh, the pure phosphorescence in the crystal may occur as a result of the irradiation during the implanting process. The authors of Ref. [11] used a Mylar foil to catch the recoil nuclei and to leave only phosphorescence. Since the Mylar foil acts on the phosphorescence, its intensity was considered to decrease by a scaling factor of 1.15. But this influence is not so trivial in the general case. As was correctly pointed in Ref. [12], the opportunity to exclude the phosphorescence with one scaling factor must be previously proved. We make this proof and, in this way, validate the method.

Electrons and holes created under radiation thermalize and then are caught by traps in the crystal. We know that the filled traps associated with radiation defects emit light for micro- and nanoseconds [16], by giving the fluorescence. Therefore, the phosphorescence must be caused by traps of another origin. We consider them to be associated with contamination centers caused by the prolonged storing of the crystal in the atmosphere. These traps stay filled for much longer times, by slowly recombining due to the following mechanism [17]. The electrons and the holes tunnel to each other under the crystal potential barrier and then recombine giving, in this way, the phosphorescence. Its intensity may be found as

$$J = \int\limits_0^{+\infty} f(r) w_T e^{-w_T t} dr, \tag{9}$$

where the integration is carried out over the distance between electrons and holes in the pairs (combinations of any electron and any hole), t is the time, $w_T = w_T(r)$ is the tunneling probability, $f(r)$ denotes the contribution of pairs at a distance r given by the expression $f(r) \propto S(r) w(r, \nu)$, where S is the overlap integral of electron's and hole's ψ-functions in the pair, and w denotes the distribution of the pairs over distances. Note that only the number w depends on the concentration of filled traps ν, which depends, in turn, on the dose of ionizing radiation. Under the assumption that the traps are scattered uniformly, the Poisson distribution gives

$$w(r, \nu) = 4\pi r^2 \nu \exp\left(-\frac{4}{3}\pi r^3 \nu\right). \tag{10}$$

The number of filled traps ν is bounded by the number of contamination centers. We consider ν to be small enough for holding $\frac{4}{3}\pi r^3 \nu \ll 1$ for such r that $S(r)$ significantly differs from zero, i.e., electron's and hole's ψ-functions overlap appreciably. Under this assumption, the exponent may be replaced by 1, and we easily obtain $w(r, \nu) \propto \nu$ and $J = \nu F(t)$, where the function F does not depend on ν. We see that a variation of ν just acts on the magnitude, but does not change the qualitative temporal behavior of the phosphorescence signal. The result is that *if the crystal is contaminated not much, the phosphorescence may be really excluded with one scaling factor.*

4. Fluorescence

There may be another appearance of ^{233}U daughter nuclei: their α- and β-particles may cause the excitation of the medium and the further fluorescence. This parasitic signal is the most difficult for the study, which must be carried out for each material of a crystal plate separately. In this paper, we consider the effect for MgF_2 and extend then the conclusions over some other materials.

We start from the fluorescence caused by β-particles. We know that it is selective by wavelength and, in our case, is significant at wavelengths $\lambda > > 225$ nm [18]. Consequently, the excluding rule reliable for Cherenkov radiation gives no information now. But the fluorescence at $\lambda > 225$ nm turns out to be of the same order as Cherenkov radiation, so they appear only together. Analyzing Ref. [18], we conclude that the β-fluorescence efficiency may be roughly estimated by the value of $\varepsilon_f = 2$ photons irradiated per 10 nm of the spectrum into a unit solid angle for the loss of electron's energy at 1 MeV.

As for Cherenkov radiation, we have already obtained the number of photons irradiated in the range

115–320 nm per an electron decelerating from the initial kinetic energy to the Cherenkov effect threshold equal to 170 keV in MgF_2. The results are shown in Table 2. For the subsequent estimations, we use the value $n = 110$ photons/MeV as the mean number of photons per lost energy 1 MeV. We now easily evaluate the "efficiency" of Cherenkov radiation $\varepsilon_c = 0.4$ photons irradiated per 10 nm of the spectrum into a unit solid angle for the loss of electron's energy at 1 MeV. We see that ε_f and ε_c differ just by 5 times, i.e., they are comparable. So, *if there is no Cherenkov radiation at all, then there is no β-fluorescence as well.* This works for almost all materials listed in Table II in Ref. [18]. This fails for CaF_2 and BaF_2, because their fluorescence efficiencies are by the order greater and cannot be compared with the Cherenkov radiation efficiency. This fact complicates the usage of these materials for nuclear recoil experiments.

If there is no Cherenkov radiation and β-fluorescence, and the phosphorescence is excluded, then only the α-fluorescence remains. To realize whether the resulting signal represents the α-fluorescence or something else, one should consider its time dependence and compare it with the time evolution of the total α-activity in the crystal plate looking for similar features. For example, as the authors of Ref. [12] pointed, the α-activity time dependence has a feature consisting in the first decrease because of the fast decay of ^{221}Fr (half-life 4.9 min) and the building up over several days (half-lives of ^{225}Ra and ^{225}Ac).

Resuming the obtained results, we see that there was no Cherenkov radiation in the experiment in Ref. [11]. Moreover, as MgF_2 was used, there could be the just little β-fluorescence. The phosphorescence was excluded with the scaling factor. The resulting signal has nothing like the first decrease and the subsequent building up during a long time. Thus, we conclude that the authors of Ref. [11] might really observe ^{229m}Th decay photons.

5. Conclusion

Applying the method of recoil nuclei, one should use uranium sources fresh enough to exclude the Cherenkov radiation and the fluorescence in a crystal used for the collection of thorium recoil nuclei. We showed that one may neglect the Cherenkov radiation caused by the β-decaying daughter nuclei of ^{233}U, if the sample was purified at the earliest 100 days ago. We deduced that if the investigated range of energies has a subrange with no PMT signal, then there is no Cherenkov radiation at all. Using these results, we conclude that the authors of Ref. [11] really did not deal with Cherenkov radiation. Moreover, we have shown that the phosphorescence in this experiment was excluded correctly. Thus, if there were no ^{229m}Th decay photons, then the resulting signal must represent only the fluorescence. But, under the conditions in Ref. [11], the time dependence of the signal is not characteristic of the fluorescence, so the authors really might observe ^{229m}Th decay photons. We carried out the necessary theoretical foundations for the method of recoil nuclei and hope for that the other experiments within this method will be carried out and give the necessary statistics for making the final conclusion on the isomer state of ^{229}Th.

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АНАЛІЗ ПАРАЗИТНИХ СИГНАЛІВ
У ЕКСПЕРИМЕНТІ ПРЯМОГО СПОСТЕРЕЖЕННЯ
ІЗОМЕРНОГО СТАНУ ^{229m}Th МЕТОДОМ
ЯДЕР ВІДДАЧІ

Р е з ю м е

Виконано необхідний теоретичний аналіз методу ядер віддачі в застосуванні до прямого спостереження ізомерного стану ^{229m}Th. Розглядається черенковське випромінювання, фосфоресценція та флуоресценція в кристалічній пластині, що використовується в ролі колектора ядер віддачі. Обговорюються шляхи уникнення паразитних сигналів для успішного спостереження фотонів розпаду стану ^{229m}Th.

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АНАЛИЗ ПАРАЗИТНЫХ СИГНАЛОВ
В ЭКСПЕРИМЕНТЕ ПРЯМОГО НАБЛЮДЕНИЯ
ИЗОМЕРНОГО СОСТОЯНИЯ ^{229m}Th
МЕТОДОМ ЯДЕР ОТДАЧИ

Р е з ю м е

Выполнен необходимый теоретический анализ метода ядер отдачи в применении к прямому наблюдению изомерного состояния ^{229m}Th. Рассматривается черенковское излучение, фосфоресценция и флуоресценция в кристаллической пластине, которая используется в качестве коллектора ядер отдачи. Обсуждаются пути предотвращения паразитных сигналов для успешного наблюдения фотонов распада состояния ^{229m}Th.