

Decay of the low-energy nuclear isomer $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ in solids (dielectrics and metals): A new scheme of experimental research

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The decay channels and the half-life of the proposed anomalously low-energy nuclear isomer $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ in a dielectric and in metals are discussed. The preferred decay channel in wide energy-gap dielectrics is via nuclear γ emission in the optical range. The isomer's half-life lies in the range 10 min–1 h in $^{229}\text{ThO}_2$ for the spectral range $\omega_N = 4.5\text{--}2.5 \text{ eV}$. Nonradiative decay channels dominate in metals. A two-step experimental scheme is developed. The first step is the excitation of the low-energy isomeric level by synchrotron radiation via the 29- and 72-keV levels, and precise measurement of the wave length of the low-energy nuclear transition in $^{229}\text{ThO}_2$. The second step is the excitation of a large number of the isomeric nuclei $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ from the ground state by laser radiation in thorium dioxide, and investigation of the α decay of the isomeric level.

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I. INTRODUCTION

Experimental data obtained by Reich and Helmer at the National Engineering Laboratory in Idaho (USA) from 1989 to 1993 [1,2] provide evidence for the existence of a level with an extremely low energy of $3.5 \pm 1.0 \text{ eV}$ in the ^{229}Th nucleus. This level corresponds to the head of the $3/2^+$ [631] rotational band [2].

The anomalously low energy of the isomeric level in ^{229}Th has generated considerable interest, because such a level should have some unusual properties. There is, for example, an optical γ ray and an electron bridge mechanism [3]. The electron bridge raises the possibility of nuclear excitation by optical photons [4] and by surface plasmons [5] and can thus lead to changes of the alpha decay rate of the ^{229}Th via the $3/2^+$ (3.5 eV) level under low-intensity laser radiation [6].

Four research teams have published results of optical measurements on ^{229}Th in the last two years. A photon emission in the optical range was observed first by Irwin and Kim [7] in 1997 and by Richardson *et al.* [8] in 1998 in a sample of ^{233}U . (^{233}U is used because the low-energy isomeric level in the ^{229}Th is populated approximately 2% of the time in the α decay of ^{233}U .) They found two peaks: visible photons with the energies 2.3–2.5 eV and UV photons with energies $\approx 3.7 \text{ eV}$. This optical emission was construed as being the isomeric nuclear transition between the low-energy isomeric level and the ground state of the ^{229}Th . The 3.7-eV photons were identified as a direct nuclear γ emission, and the 2.3–2.5-eV photons as an inelastic electronic bridge. The new

range $3.5 \pm 0.5 \text{ eV}$ for the isomer's energy was obtained in Ref. [8].

The optical spectra obtained in Refs. [7,8] were revised by two research teams in 1999. Utter *et al.* [9] and Shaw *et al.* [10] proved that the optical emission with photon energy in the range near 3.7 eV corresponds to emission from nitrogen induced by α particles in the air that surrounds the radioactive ^{233}U sample. The origin of the optical emission in the range 2.3–2.5 eV was still unclear.

A line at 391.3 nm ($\approx 3.17 \text{ eV}$) was found by Shaw *et al.* [10] in the optical emission spectrum of ^{233}U in air. This line was absent in the emission spectrum of an atmospheric-pressure N_2 discharge lamp (with a 3-kV applied voltage), which was used in the work [10] for comparison of spectra. The 391.3-nm line, however, is very close to the known N_2^+ line at 391.4 nm [11,9]. The fluorescence of nitrogen under the influence of the 5-MeV α particles probably differs from the fluorescence of nitrogen inside the 3-kV discharge lamp.

In their latest article, Young *et al.* [12] established a plausible origin for the 2.5-eV optical emission in the $^{233}\text{UO}_2(\text{NO}_3)_2$ solution and in the $^{233}\text{UO}_4 \cdot 2\text{H}_2\text{O}$ sample placed in a silica tube. This emission was thought to be at least in part due to fluorescence of the uranyl ions excited by the α radioactivity of the $^{233}\text{UO}_2(\text{NO}_3)_2$ solution, and due to fluorescence of the silica tube caused also by the α radioactivity.

The results obtained Refs. [7–10,12] demonstrate that a judicious selection of the chemical compound counts for a great deal in determining the $3/2^+$ ($3.5 \pm 1.0 \text{ eV}$) level properties by optical methods. Therefore one should know how this level decays in different chemical compounds, and how these chemical compounds behave under the action of α particles, x rays, optical photons, and electrons.

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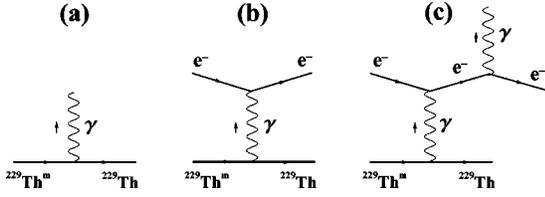


FIG. 1. Diagrams of the $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ decay. (a) Nuclear γ radiation in the optical range, (b) inelastic scattering of conduction electrons by the nuclei and internal conversion at the valence band electrons, (c) electronic bridge process.

Thorium and uranium are reactive metals. They form chemical compounds with various widely differing physical properties. The decay channels of the $3/2^+(3.5 \pm 1.0 \text{ eV})$ level depend strongly on electronic properties of these chemical compounds. That is why we consider here first how the level decays in some chemical compounds with known electronic properties, such as dielectrics and metals.¹ After that, we propose a new experimental method for detection of the $3/2^+(3.5 \pm 1.0 \text{ eV})$ level.

II. DECAY OF $^{229}\text{Th}^m(3/2^+, 3.5 \pm 0.1 \text{ eV})$ IN A DIELECTRIC

Thorium dioxide ThO_2 is the most accessible and stable among the thorium chemical compounds. ThO_2 is a wide energy-gap solid dielectric: thorium dioxide has an $\approx 6\text{-eV}$ insulating gap [13,14]. Let us consider the decay channels of the $3/2^+(3.5 \pm 1.0 \text{ eV})$ nuclear level in the dielectric $^{229}\text{ThO}_2$.

All possible decay processes of the $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$, except for the α decay, are shown in Fig. 1. It is evident that internal conversion [Fig. 1(b)] is forbidden energetically in the $^{229}\text{ThO}_2$, because the value of the insulating gap exceeds the nuclear transition energy. (Internal conversion is a process of electron ejection from the valence band to the conduction band.)

The electronic bridge process is shown in Fig. 1(c). There are no electronic states between the valence and conducting bands in a pure dielectric. As a result, only a nonresonant elastic electronic bridge via the virtual intermediate electronic states from the conducting band is possible. The difference between the nuclear transition energy and energies of the excited electronic states in the conduction band is large, more than 1 eV. Furthermore, both virtual and real photons in Fig. 1(c) have $M1$ multiplicities in the elastic electronic bridge considered here for decay of the $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$. Such an electronic bridge process has a negligibly small probability [3].

Nuclear γ emission in the optical range [this process is shown in Fig. 1(a)] is possible in ThO_2 . The probability of $M1$ emission can be estimated from the formula [15] (adopted system of units is $\hbar = c = 1$)

$$W_{\gamma}^{M1} = 10n^3 \omega_N^3 \mu_N^2 B_{\text{W.u.}}(M1), \quad (1)$$

¹Decay of the $^{229}\text{Th}^m(3.5 \pm 1.0 \text{ eV})$ in semimetals and semiconductors will be considered in detail in a separate article.

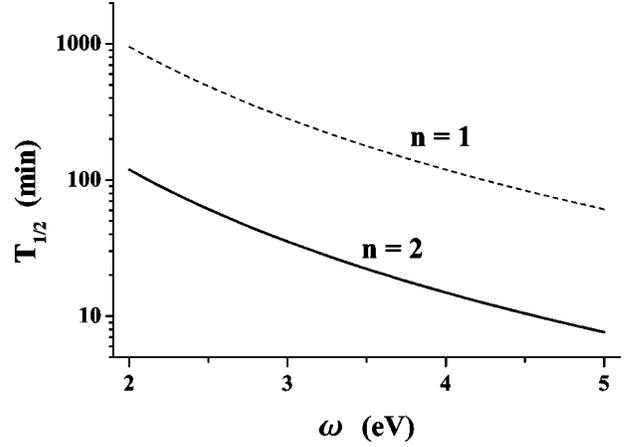


FIG. 2. Half-life of $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ in the dielectric $^{229}\text{ThO}_2$ (the refractive index $n=2$), and in vacuum ($n=1$), if the nuclei decay via the first-order process in Fig. 1(a).

where $\mu_N = e/2M$ is the Bohr magneton, e and M are the proton charge and mass correspondingly, $B_{\text{W.u.}}$ is the nuclear reduced probability in Weisskopf units, ω_N is the energy of the nuclear transition, n is a refractive index ($n=1$ in vacuum, and $n=2$ in the thorium dioxide [14]).

The reduced probability of the nuclear $M1$ low-energy isomeric transition in ^{229}Th has been calculated [16]. The value

$$B_{\text{W.u.}}[M1; 3/2^+(3.5 \pm 1.0 \text{ eV}) \rightarrow 5/2^+(0.0)] \approx 4.8 \times 10^{-2} \quad (2)$$

was determined taking into account the Coriolis mixing of the ground state rotational band $K^\pi[Nn_Z\Lambda] = 5/2^+[633]$ and of the low-energy isomeric state rotational band $3/2^+[631]$.

Under the abovementioned conditions, one obtains for the half-life $T_{1/2}^{is}$ of the $3/2^+(3.5 \pm 1.0 \text{ eV})$ isomeric level in thorium dioxide the values shown in Fig. 2. It should be noted, this is the first known case where the probability of a nuclear transition depends strongly on the index of refraction. As a result, the time range $T_{1/2}^{is} \approx 10 \text{ min} - 1 \text{ h}$ corresponds to the energy range $\omega_N = 4.5 - 2.5 \text{ eV}$ for the $3/2^+(3.5 \pm 1.0 \text{ eV})$ level decay in thorium dioxide.

III. DECAY OF THE $^{229}\text{Th}^m(3/2^+, 3.5 \pm 0.1 \text{ eV})$ IN A METAL

The critically important result of Utter *et al.* [9] shows a total disappearance of the optical emission from the ^{233}U sample in vacuum. Let us consider one possible reason for the negative result of the vacuum measurements. The sample of ^{233}U in the experiment [9] was prepared by electroplating a thin layer of uranium on a platinum disk. This contrasts with works [7,8,10] where a metal substrate was not used. Isomeric nuclei $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ arise from the α decay of ^{233}U . If the ^{233}U nuclei are placed on the surface of a Pt plate, there are, in principle, three different possibilities for the recoil nuclei. They can stop in the ^{233}U sample itself, or they can fly into the chamber, or embed themselves in the Pt substrate.

The kinetic energy of the recoil nuclei is ~ 100 keV. Their track length is ~ 200 Å in the $^{233}\text{UO}_2$ sample [17]. That is why a few recoil nuclei ($\sim 1\%$) stopped in the sample itself in the experiment [9] if the $^{233}\text{UO}_2$ thickness was 35 Å [18].

Consider first the case where the $^{229}\text{Th}^m$ nuclei move into the vacuum chamber. The velocity of the recoil nuclei is $\sim 10^{-3}$. The nuclei transit time in the chamber is very small (less than 10^{-6} s). The half-life of the low-energy isomer exceeds this time (see Ref. [16]), and the isomeric nuclear decay occurs at the chamber walls, beyond the field of vision of the optical system.²

The situation, when the $^{229}\text{Th}^m$ nuclei move to the Pt plate, is more interesting than the previous one. Recoil nuclei with 100-keV energies penetrate the metal plate to a depth of some tens of atomic layers ($\sim 100\text{--}150$ Å [17]). Such a layer of metal is transparent to optical photons. But, the decay modes of the low-energy isomeric level $3/2^+$ (3.5 ± 1.0 eV) inside a metal can differ from the usual decay modes. It is known that internal conversion is forbidden energetically in the Th atom [3]. And a third-order process, the electron bridge, is the main decay channel of the $^{229}\text{Th}^m$ in an isolated atom [3]. That is why a more important difference for the $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0$ eV) decay in a metal is the following. These isomers can decay via conduction electrons [19] [this virtual one-photon process in Fig. 1(b) does not have a reaction threshold], and via electrons from the valence band (this process is possible if ω_N exceeds the Fermi energy).

The decay via conduction electrons is the inverse of the process of inelastic electron scattering on nuclei and is shown in Fig. 1(b). (One can view this process as the analog of internal conversion for the conduction electrons.) The energies of the initial (E_i) and the final (E_f) electron states are connected by the formula $E_f = E_i + \omega_N$.

The probability of decay is calculated from the formula

$$W_{e,e'} \sim N_e v_e \sigma_{e,e'}, \quad (3)$$

where N_e is the density of conduction electrons, v_e is the velocity of electrons, $\sigma_{e,e'}$ is the cross section for the process.

One can use a simple model of an electron gas in metal [20] for a qualitative estimation of $W_{e,e'}$. Let us take $E_i \approx \mathcal{E}_F$, $v_e \sim v_F = \sqrt{2\mathcal{E}_F/m}$, where v_F and \mathcal{E}_F are the Fermi velocity and Fermi energy correspondingly, m is the electron mass.

²It should be noted that the radiative relaxation time of the Th atomic shell after the ^{233}U α decay does not exceed the typical time for optical transitions in the Th atomic shell $\sim 10^{-8}$ s. The recoil nuclei are within a radius of 1 cm of the sample during this interval of time, and relaxation of the Th atomic shell can be detected by the optical system. However, the optical system in the experiment [9] did not detect an optical emission in vacuum. This means that optical photons were not emitted at all in the process of relaxation of the Th atomic shell after the ^{233}U α decay in this experiment [9].

Suppose a metal is on the wall of the vacuum. If the value of the electronic work function of the metal is bigger than ω_N , conduction electrons remain in the metal after they absorb the energy ω_N at the inelastic scattering by the $^{229}\text{Th}^m$ nuclei. (If a metal is on the wall of uranium dioxide or some other material, one should know the effective electronic work function of this complex.) After that, the excitation energy transforms into heat over the relaxation time $\sim 10^{-14}$ s [20].

Calculation of the cross section $\sigma_{e,e'}$ was based on the set of computer programs developed by Band *et al.* [21]. The electron wave functions of binding states for a self-consistent atomic field are obtained by solving the relativistic Dirac-Fock equations taking into account the finite nuclear size.

Under these assumed conditions one obtains for the cross section $\sigma_{e,e'} \sim 10^{-27}$ cm² with the reduced probability of the nuclear $M1$ transition $B(M1; 5/2^+ 5/2[633] \rightarrow 3/2^+ 3/2[631])_{\text{w.u.}} \approx 3.2 \times 10^{-2}$, which follows from Eq. (2).

Using the values $N_e \approx 6 \times 10^{22}$ cm⁻³ and $\mathcal{E}_F \approx 5.5$ eV for ‘‘standard’’ metal [22], one can estimate a decay probability of the $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0$ eV) inside a metal substrate. From Eq. (3) it follows that $W_{e,e'} \sim 10^4$ s⁻¹, i.e., the isomer’s half-life in a metal is $\sim 10^{-4}$ s. This time is considerably smaller than the lower bound for the half-life of the isomer $\sim 10^{-2}$ s established theoretically in the work [16] as a ‘‘reasonable lower bound.’’ Therefore the ^{229}Th low-energy isomers can decay inside a metal substrate without photon emission.

The second nonradiative decay channel of $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0$ eV) in a metal is via internal conversion of the valence-band electrons. All electronic states below the Fermi energy are occupied in a metal. That is why internal conversion is allowed, if the nuclear transition energy exceeds the Fermi energy. In this case, valence electrons of the Th atom are ejected into the conducting band. One can use a model of an electron gas in a metal again for the qualitative estimation of the probability W_{conv} for this process.

Calculations of W_{conv} have been described in detail [3]. We used a set of computer programs from Ref. [21]. The valence atomic shell of the Th atom is $6d^2 7s^2$ [23]. $W_{\text{conv}} \sim 10^6$ s⁻¹ if $7s$ electrons are ejected into the conducting band, and $\sim 10^3$ s⁻¹ if $6d$ electrons absorb the $M1$ virtual photon emitted by the nucleus. This is a qualitative estimate only. W_{conv} depends on the structure of the conducting band, and on effects of the multiple scattering of low-energy electron by neighboring atoms. Such effects are well known in XANES (x-ray absorption near edge structure [24]). They can change the value of W_{conv} by orders of magnitude [24]. This means that thorium atoms with nuclei $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0$ eV) could be used as a sensitive probe for the investigation of properties of materials.

IV. EXCITATION OF THE $^{229}\text{Th}^m(3/2^+, 3.5 \pm 0.1$ eV) BY SYNCHROTRON AND LASER RADIATION

The dielectric $^{229}\text{ThO}_2$ is a more suitable chemical compound for a first experiment. This follows from the results of

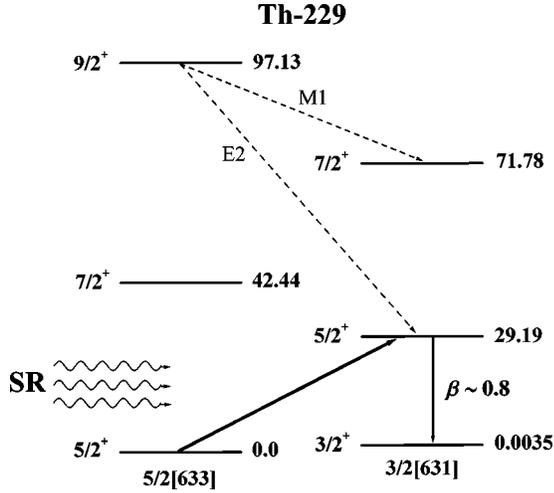


FIG. 3. Excitation of the $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ level by Synchrotron Radiation (SR) via the intermediate level $5/2^+(29.19 \text{ keV})$. Dashed lines are the $M1$ and $E2$ nuclear transitions measured in Ref. [25].

Sec. II. Radiative decay of the $3/2^+(3.5 \pm 1.0 \text{ eV})$ level is shown in Fig. 1(a). This process occurs in the thorium dioxide with emission of a nuclear optical photon, and gives us the possibility to measure a wave length of the nuclear transition.

For the optical measurements one should first excite a large number of the isomeric nuclei $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$. And second, the excitation method should not break the dielectric structure of the $^{229}\text{ThO}_2$ sample (i.e., should not create luminescence centers, for example). One can achieve this by using synchrotron radiation (see Fig. 3). The isomeric level $3/2^+(3.5 \pm 1.0 \text{ eV})$ can be excited via the intermediate nuclear levels $5/2^+(29.19 \text{ keV})$ and $7/2^+(71.78 \text{ keV})$ by monochromatic 29.19 keV and 71.78 keV synchrotron radiation, correspondingly.

The low-energy levels of a ground-state rotational band $K^\pi[Nn_z\Lambda] = 5/2^+[633]$ and the $3/2^+[631]$ band in ^{229}Th are displayed in Fig. 3. The interband $M1$ $9/2^+(97.13 \text{ keV}) \rightarrow 7/2^+(71.78 \text{ keV})$ transition, and $E2$ $9/2^+(97.13 \text{ keV}) \rightarrow 5/2^+(29.19 \text{ keV})$ transition have been measured [25]. Their reduced transition probabilities are $B_{\text{W.u.}}[M1; 9/2^+(97.13 \text{ keV}) \rightarrow 7/2^+(71.78 \text{ keV})] \approx 0.018$, $B_{\text{W.u.}}[E2; 9/2^+(97.13 \text{ keV}) \rightarrow 5/2^+(29.19 \text{ keV})] \approx 5$. The Coriolis interaction of rotational bands $5/2^+[633]$ and $3/2^+[631]$ is relatively small in the ^{229}Th [25,26], and one can use the Alaga pure rotation relations for the calculation of $M1$ and $E2$ reduced transition probabilities between the levels of these bands.

The radiative widths for excitation of the level $5/2^+(29.19 \text{ keV})$ from the ground state are $\Gamma^{\text{rad}}[M1; 5/2^+(0.0) \rightarrow 5/2^+(29.19 \text{ keV})] \approx 5.6 \times 10^{-9} \text{ eV}$, $\Gamma^{\text{rad}}[E2; 5/2^+(0.0) \rightarrow 5/2^+(29.19 \text{ keV})] \approx 0.92 \times 10^{-11} \text{ eV}$. These values were obtained with reduced transition probabilities $B_{\text{W.u.}}[M1; 5/2^+(0.0) \rightarrow 5/2^+(29.19 \text{ keV})] \approx 0.011$, $B_{\text{W.u.}}[E2; 5/2^+(0.0) \rightarrow 5/2^+(29.19 \text{ keV})] \approx 6.5$.

For estimation of the branching ratio $\beta[5/2^+(29.19 \text{ keV}) \rightarrow 3/2^+(3.5 \text{ eV})]$, we calculated the

conversion coefficients for the transition energy 29.19 keV: $\alpha_{M1} \approx 158$, $\alpha_{E2} \approx 4683$. The reduced transition probability of the intraband $M1$ transition between $5/2^+(29.19 \text{ keV})$ and $3/2^+(3.5 \text{ eV})$ levels $B_{\text{W.u.}}[M1; 5/2^+(29.19 \text{ keV}) \rightarrow 3/2^+(3.5 \text{ eV})] \approx 0.027$ was calculated with a value $|g_K - g_R| = 0.58$ [26], where g_R and g_K are rotational and intrinsic gyromagnetic ratios, correspondingly. We estimated the reduced transition probability $B_{\text{W.u.}}[E2; 5/2^+(29.19 \text{ keV}) \rightarrow 3/2^+(3.5 \text{ eV})] \approx 280$ using for the intrinsic quadrupole moment Q_{20} the value $8.816e \text{ b}$ [25], measured for the ground-state rotational band. As a result, we obtained for $\beta[5/2^+(29.19 \text{ keV}) \rightarrow 3/2^+(3.5 \text{ eV})] \approx 0.8$. [Our estimation of the $5/2^+(29.19 \text{ keV})$ level half-life is $\sim 0.1 \text{ ns}$].

As an example we evaluate a number of the excited nuclei $^{229}\text{Th}^m(3/2^+, 3.5 \text{ eV})$ for the case of synchrotron radiation of the Advanced Photon Source (APS) at Argonne National Laboratory. The APS has the following key parameters: the beam current $I = 100\text{--}300 \text{ mA}$, the beam energy $E = 7 \text{ GeV}$, the critical energy $\omega_C = 32.6 \text{ keV}$ [27]. It is easy to estimate a spectral flux of photons with energy $\omega = 29.19 \text{ keV}$ in the photon energy interval $\Delta\omega = \Gamma^{\text{rad}} = 5.6 \times 10^{-9} \text{ eV}$ in 1 mrad of horizontal angle by the formula [28]

$$N(\omega) (\text{photons/s mrad}) = 2.46 \times 10^{13} I (\text{mA}) E (\text{GeV}) \eta(\omega/\omega_C) \frac{\Delta\omega}{\omega}, \quad (4)$$

where $\eta(\omega/\omega_C)$ is a universal synchrotron radiation spectrum. The function $N(\omega)$ in formula (4) is integrated over the vertical angle.

For the $10 \mu\text{g}$ $^{229}\text{ThO}_2$ sample (the number of thorium nuclei is $N_{\text{Th}} \approx 2.3 \times 10^{16}$) placed at a distance of 15 m from a synchrotron radiation source at the plate $0.5 \times 0.5 \text{ cm}^2$, the excitation rate of the $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ is $\sim 10^3 \text{ nuclei} \times \text{s}^{-1}$. This means that the optical activity of the sample will be $\sim 10^3 \text{ s}^{-1}$ after the $\sim 1 \text{ h}$ irradiation. As the calculation shows, excitation of the $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ via the level $7/2^+(71.78 \text{ keV})$ by synchrotron radiation is possible too, but it gives an excitation rate smaller by an order of magnitude approximately.

Excitation of the $3/2^+(3.5 \pm 1.0 \text{ eV})$ level by monochromatic 29.19 keV synchrotron radiation via the $5/2^+(29.19 \text{ keV})$ intermediate level enable us to avoid excitation of molecular levels, and excitation of luminescence centers inside the solid sample. Monochromatic 29.19 keV synchrotron radiation does not damage much the dielectric structure of $^{229}\text{ThO}_2$ sample. For example, the optical activity $\sim 10^3 \text{ s}^{-1}$ of the $^{229}\text{ThO}_2$ sample can be obtained under the same conditions after irradiation by a 10-mA electron beam with 30–40 keV energy electrons. However, electrons can destroy the dielectric structure of the sample. They can create a lot of luminescence centers, and so on. That is why a synchrotron radiation seems to be a more applicable instrument for such an experiment.

The optical activity $\sim 10^3 \text{ s}^{-1}$ is enough for measurement of the wave length of the low-energy $3/2^+(3.5 \pm 1.0 \text{ eV}) \rightarrow 5/2^+(0.0)$ nuclear isomeric transition. After this one can use a tunable laser for photoexcitation of the

$3/2^+(3.5 \pm 1.0 \text{ eV})$ level from the ground state in thorium dioxide $^{229}\text{ThO}_2$. The radiative width of this nuclear transition is $\Gamma^{\text{rad}}[5/2^+(0.0) \rightarrow 3/2^+(3.5 \text{ eV})] \sim 10^{-19} \text{ eV}$ according to the results of Sec. II. The resonance cross section can be evaluated from the formula

$$\sigma_{\text{res}} \approx \frac{\lambda_N^2}{2\pi} \frac{\Gamma^{\text{rad}}[5/2^+(0.0) \rightarrow 3/2^+(3.5 \text{ eV})]}{\Delta\omega_L},$$

where $\lambda_N = 2\pi/\omega_N$, $\Delta\omega_L$ is the laser line width. The cross section is $\sigma_{\text{res}} \sim 10^{-25} \text{ cm}^2$ for a typical tunable laser beam with the ratio $\Delta\omega_L/\omega_L \sim 10^{-5}$ in the range $\omega_L = \omega_N = 3.5 \pm 1.0 \text{ eV}$ [4]. The number of isomeric nuclei excited per unit time is given by

$$\mathcal{N}_i \sim N_{\text{Th}} \sigma_{\text{res}} \phi_L, \quad (5)$$

where ϕ_L is the flux density of the laser photons. One can excite $\sim 10^{10}$ isomeric nuclei $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ per second by a 1-W power laser in the thorium dioxide sample described above. The number of isomeric nuclei that can be obtained after a ~ 1 -h irradiation, is about 10^{12} – 10^{13} (depending on the isomeric level half-life). Thus, this gives us a chance to find the α decay of the isomeric level $3/2^+(3.5 \pm 1.0 \text{ eV})$, and to obtain independent evidence on the existence of this level.

According to Ref. [6], the isomeric level α decay constant λ_{is}^α and the α decay constant of the ground state

λ_{gr}^α ($\approx 2.8 \times 10^{-12} \text{ s}^{-1}$) obey the relation $2 \leq \lambda_{is}^\alpha / \lambda_{gr}^\alpha \leq 4$. The α spectrum emitted by the ^{229}Th nuclei will change, if the isomeric nuclei $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ are present in the sample. The 4.930 MeV line is absent practically in the α spectrum of the ^{229}Th (the intensity of this line is 1.6×10^{-3} per α decay of the ^{229}Th ground state [29]). Under laser radiation this 4.930 MeV α line will appear. This effect can be detected by measuring the α spectrum, or by measuring the intensity of some γ rays. The population of the $3/2^+(149.96 \text{ keV})$ level in the daughter nuclei ^{225}Ra increases as a consequence of the direct α decay of the isomeric nuclei $^{229}\text{Th}^m(3/2^+, 3.5 \pm 1.0 \text{ eV})$ to the $3/2^+(149.96 \text{ keV})$ state in the ^{225}Ra . The intensities of the γ rays from this state will increase. The energies of these γ rays are 94.73 keV, 107.108 keV, 124.55 keV, and 150.04 keV [29]. Without such evidence, one cannot be fully convinced that the $3/2^+(3.5 \pm 1.0 \text{ eV})$ level is being detected.

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