Decay channel of low-lying isomer state of the ^{229}Th nucleus. Possibilities of experimental investigation

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Possible decay channels for low-lying ($E_{i.s.} \leqslant 5$ eV) isomer states of the thorium-229 nucleus (internal electron conversion, γ-quantum emission by the nucleus, electronic bridge) are discussed on the basis of theoretical calculations. The probabilities of these processes as functions of $E_{i.s.}$ are calculated. An experimental setup for measuring the isomer energy and lifetime is proposed.

1. INTRODUCTION

The thorium-229 nucleus has an anomalously low-lying state with unknown half-life and energy (designated $E_{i.s.}$) estimated until recently at about ten electron volts.[1] A value 1 ± 4 eV was obtained in Ref. 2 for $E_{i.s.}$ on the basis of an analysis of gamma transitions in ^{229}Th to the ground and first excited states from high-lying energy levels populated in α-decay of uranium-233 nuclei.

Such an unusually low energy, on a nuclear scale, of a first excited state is an interesting anomaly that will undoubtedly attract the attention not only of nuclear physicists but also of specialists working in "low-energy" physics, optics, solid-state physics, lasers, plasma, and others.

Closely spaced levels with energy spacing up to several tens of electron volts are encountered quite frequently at excitation energies on the order of hundreds of electron volts. It is impossible, however, to observe transitions between them, owing to the vanishingly low probabilities against the background of decays to low-lying states. These transitions can be investigated only in isomer decays of low-lying levels. The isomer level with a lifetime on the order of 25 min had the lowest excitation energy (76.8 eV) of all the states known until recently. It decays as a result of internal electron conversion and has by now been well investigated.

Our purpose here is to estimate numerically the decay probabilities as a function of the isomer-level energy; these are needed to organize an experimental determination of the energy and lifetime of the first excited state of the ^{229}Th nucleus. We discuss three isomer-level decay channels: electron internal conversion (Sec. 2), γ-quantum emission by the nucleus, and electron bridge (Sec. 3). In Sec. 4 we consider a method of obtaining a large number of isomer thorium-229 nuclei within times comparable with the lifetime $T_{1/2}$ of the first excited level in a plasma produced, for example by laser radiation or by an electron beam.

Since the task is not to describe in detail a known experimental situation but to obtain preliminary estimates in a part of the results, namely, to determine the probabilities of the processes, we confine ourselves to order-of-magnitude estimates. We use the system of units with $\hbar = c = k = 1$.

2. INTERNAL ELECTRON CONVERSION

The upper bound 5 eV of the isomer-state energy can cause the situation with the decay channels of this level to deviate strongly from the traditional situation. According to the data of Ref. 3 the ionization potential of the thorium atom is 6.08 eV, so that a transition of the nucleus to the ground state as a result of internal electron conversion (IEC) in an isolated atom is impossible. We, however, have deemed it necessary to consider briefly here the conversion channel of the decay for the following reasons. First, the upper bound established in Ref. 2 for $E_{i.s.}$ is very close to the ionization potential (the difference is only 1.08 eV), and has been obtained only from an analysis of γ transitions of tens of kiloelectron volts and so far in only one experiment. It is evidently premature to regard this bound as absolutely reliable. Second, an independent estimate of $E_{i.s.}$ can be obtained just from a conversion experiment. By varying the chemical surroundings of the thorium atoms and changing thereby the ionization potential, it is possible to obtain a reliable estimate of the isomer-state energy. To organize such an experiment one must know the characteristic decay times of a nuclear level. It is this which we estimate here. Third, IEC from excited shells of the Th atom may turn out to be energywise allowed. By throwing electrons with the aid of laser radiation of appropriate wavelength to higher and higher excited states it will be possible to open a conversion decay channel.

Note that for certain values of $E_{i.s.}$ the probability of IEC from a number of excited states turns out theoretically to be considerably higher than from the ground state. An interesting possibility arises of altering the lifetime of an isomer level of a nucleus by action of laser radiation on the atom.

According to our calculations, in which the mean field and the electron wave functions were calculated in a relativistic variant of the Hartree–Fock–Slater method with Later's correction (the program was kindly provided by A. A. Soldatov), conversion at $6.08 \leqslant E_{i.s.} \leqslant 10$ eV is possible only from the $6D_{3/2}$ and $7S_{1/2}$ atomic shells. The conversion probability per unit time from these shells, for a nuclear transition of multipolarity $M1$ and energy ω_N, was calculated using the equation

$$W_e(M1, {}^3/_2{}^+ \to {}^5/_2{}^+, \omega_N) = 8\pi e^2 \frac{\omega_N{}^3}{9} \frac{\varepsilon+m}{p} B(M1; {}^3/_2{}^+ \to {}^5/_2{}^+) \times \sum_{l_i, j_f} (2j_f+1)\,(j_i{}^1/_2 10 | j_f{}^1/_2)^2 \Big| (\varkappa_i+\varkappa_f) \times \int_0^\infty dr\, r^2 h_1^{(1)}(\omega_N r)\,[g_i(r) F_{l_f j_f}(r) + f_i(r) G_{l_f j_f}(r)\Big|^2. \quad (1)$$

Here e and m are, respectively, the charge and mass of the

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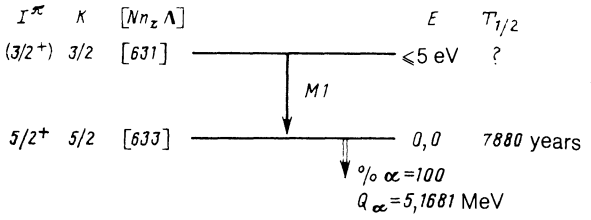

FIG. 1. Lower states of ^{239}Th nucleus. $[Nn_z\Lambda]$—quantum numbers in the Nilsson model.

electron, ε and p are its energy and momentum in the final state, $\varkappa=(l-j)(2j+1)$, l and j are the orbital and total momenta of the electron, $h_L^{(1)}$ is a Hankel function of the first kind, and $(j_i\tfrac{1}{2}10|j_f\tfrac{1}{2})$ is a Clebsch–Gordan coefficient. The wave functions of the initial state are normalized by the condition

$$\int_0^\infty [g_i^2(r)+f_i^2(r)]r^2\,dr=1,$$

and those of the final state are fixed as $r\to\infty$ by an asymptote of the form $G_{lj}(pr)\to\sin(pr+l\pi/2+\hat{\delta}_{lj})$. For the reduced probability of the $B(M1;\frac{3}{2}^+\to\frac{5}{2}^+)$ nuclear $M1$ transition we chose a value smaller by two orders of magnitude than the reduced probability $B(M1,W)$ in the Weisskopf model. This attenuation factor F_{M1} was obtained in Ref. 4 for the intensities of γ transitions in the lower part of the thorium-229 spectrum between rotational-band levels with $K=\frac{5}{2}$ and $K=\frac{3}{2}$, the bases for which are, respectively, the ground and isomeric states (see Fig. 1). It agrees well, incidentally, with the nonrigorous hindrance in the asymptotic quantum numbers of the Nilsson model.

A numerical computation, whose program is described in detail in Ref. 5, yielded the internal electron conversion probability $E_{i.s.}$ in the range from 6.1 to 10 eV. On the $6D_{3/2}$ it was approximately $0.4\cdot10^3\ s^{-1}$. Accordingly, the expected experimental lifetime of the isomer level is of the order of milliseconds. On the $7S_{1/2}$ subshell, which is approximately 1 eV lower than $6D_{3/2}$ (i.e., on which the binding energy is higher), the conversion probability is higher by almost three orders, viz., $W_c\approx0.3\cdot10^6\ s^{-1}$. The lifetime for conversion from this shell is of the order of microseconds. So steep an increase of the decay probability when $7S_{1/2}$ is "turned on" can also be used in experiment.

We present finally the computed probabilities of IEC from a number of excited states for $E_{i.s.}$ exceeding somewhat the binding energies of the electrons on these states (the IEC probabilities are practically independent of the isomer-level energy in the calculated range $|E_b|\leqslant E_{i.s.}\leqslant10$ eV (E_b is the electron binding energy), and we therefore present only single values of W_c): $7P_{1/2}-3\cdot10^3\ s^{-1}$, $7P_{3/2}-40\ s^{-1}$, $7D_{3/2}-4\ s^{-1}$, $8S_{1/2}-10^4\ s^{-1}$, $8P_{1/2}-10^3\ s^{-1}$, $8P_{3/2}-10\ s^{-1}$, $9S_{1/2}-4\cdot10^3\ s^{-1}$.

Note that the obtained isomer-decay probabilities are upper-bound estimates. The attenuation we assumed for the nuclear $M1$ transition on the 10^{-2} level is not typical of magnetic-dipole transitions in the lower parts of nuclear spectra. The transitions are usually attenuated more strongly. Some caution is therefore necessary when the obtained values of W_c are used.

3. DE-EXCITATION VIA AN ELECTRON BRIDGE

If $E_{i.s.}$ turns out to be lower than the ionization energy of the atom, internal electron conversion is energywise forbidden. Depending on the isomer-state energy, the main de-excitation channel is then either direct γ radiation or an electronic bridge (EB), which is a process of third order in the electromagnetic-interaction constant (Fig. 2).

Processes with EB were observed experimentally for the nuclei ^{93}Nb (Ref. 6) and ^{193}Ir (Ref. 7) in the decay of isomer states with energies 30.7 and 80.27 keV, respectively. The probability of de-excitation through an electronic bridge ($W^{(3)}{}_\gamma$) relative to the probability of direct γ radiation ($W^{(1)}{}_\gamma$) was 7% in the first case and 21% in the second. In both cases, however, the main channel of the isomer $M4$ transition is internal electron conversion. $W^{(3)}{}_\gamma$ is very small with respect to it—approximately $1.16\cdot10^{-6}$ in ^{93}Nb and $0.95\cdot10^{-5}$ in ^{293}Ir.

Transitions via EB were investigated theoretically in Refs. 8–10 for a nuclear isomer transition energy significantly higher than the binding energy of the electrons participating in the process, and also in Refs. 11 and 12 for the decay of the low-lying (76.8 eV) isomer level in ^{235}U. In contrast to ^{93}Nb and ^{193}Ir, according to Refs. 11 and 12 the EB probability in the decay of ^{235}U exceeds the emission probability of the nucleus by several orders. Here too, however, in view of the very high conversion probability (with a coefficient of order 10^{20}) it will be difficult to distinguish a transition via an electronic bridge from the conversion background. The situation in thorium-229 is apparently entirely different.

To calculate the EB probability we introduce a number of simplifications. First, we consider only the direct diagram. A numerical computation has shown that, just as in other known cases,[10] the contribution of the exchange diagram to the process is small. Second, we shall use the single-level approximation. It is justified in this case not by the proximity of $E_{i.s.}$ to the energy of some arbitrary atomic transition (the entire range of which is only 5 eV), but by the structure of the atomic wave functions connected by EB. For

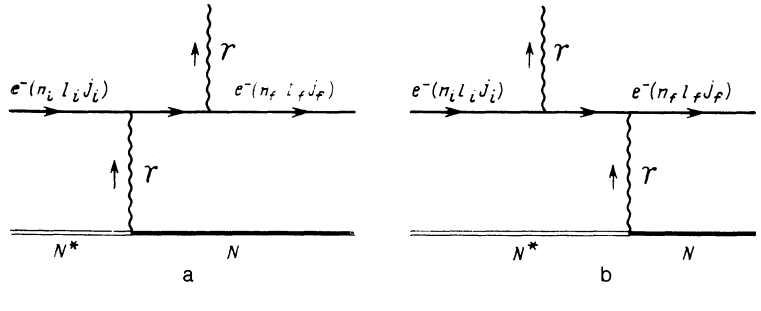

FIG. 2. De-excitation of a nucleus via an electron bridge: a—direct diagram, b—exchange diagram.

the states $6D_{3/2}$ and $7S_{1/2}$, from which an effective EB is possible in the atomic shell theory at $E_{i.s.} \leqslant 5$ eV, there exists one structure—two pairs of levels coupled by an electron bridge, and the probability of a transition through them is several orders higher than the probability of an EB through other levels. Therefore allowance for the corresponding amplitudes does not alter the results qualitatively.

Just as in Refs. 10 and 11, we introduce and calculate the ratio of the probabilities $W_\gamma^{(3)}$ and $W_\gamma^{(1)}$, which does not depend on the nuclear matrix element:

$$\zeta = \frac{W_\gamma^{(3)}}{W_\gamma^{(1)}} = \frac{\alpha_d(i \to n; E(M)L; \omega_N)\Gamma_\gamma(n \to f; E(M)L'; \omega)}{2\pi[(E_{i.s.} - E_b^{(n)} + E_b^{(i)})^2 + \Gamma_n^2/4]}, \quad (2)$$

where E_b is the binding energy of the atomic states (i—initial, n—intermediate, f—final), $\omega = E_b^{(n)} - E_b^{(f)}$, Γ_n is the total width of the intermediate atomic state, α_d is a "discrete conversion coefficient" introduced in Ref. 11 (it has the dimension of energy and is some analog of the usual internal-conversion coefficient, but for transition of an electron between states of a discrete spectrum, Γ_γ the radiative width of an atomic transition of multipolarity L' with energy ω from state n to f, and L is the multipolarity of the nuclear isomer transition. Using the corresponding equations of Refs. 13 and 14 it is easy to obtain the necessary equations for α_d and Γ_γ:

$$\alpha_d(i \to n; E(M)L; \omega_N) = \pi e^2 \omega_N \frac{2J_i+1}{L(L+1)} (j_i^{1}/_{2} L 0 | j_n^{1}/_{2})^2 |\mathfrak{M}_{E(M)L}(\omega_N)|^2, \quad (3)$$

where

$$\mathfrak{M}_{EL}(\omega) = \int_0^\infty dr\, r^2 \{h_L^{(1)}(\omega r) L[g_i(r)g_n(r) + f_i(r)f_n(r)] + h_{L-1}^{(1)}(\omega r)[(\varkappa_i + \varkappa_n - L)g_i(r)f_n(r) + (\varkappa_i + \varkappa_n + L)f_i(r)g_n(r)]\},$$

$$\mathfrak{M}_{ML}(\omega) = (\varkappa_i + \varkappa_n) \int_0^\infty dr\, r^2 h_L^{(1)}(\omega r)[f_i(r)g_n(r) + g_i(r)f_n(r)],$$

and

$$\Gamma_\gamma(n \to f; E(M)L'; \omega) = 2e^2\omega \frac{2L'+1}{L'(L'+1)} (j_n^{1}/_{2} L 0 | j_f^{1}/_{2})^2 |\mathfrak{M}'_{E(M)L'}(\omega)|^2, \quad (4)$$

where $\mathfrak{M}'_{E(M)L}(\omega) = \mathrm{Re}\{\mathfrak{M}_{E(M)L}(\omega)\}$. All the wave functions of the discrete spectrum are normalized by the same condition as in Sec. 2.

Calculation using Eqs. (2)–(4) has shown that ζ is less than unity and, accordingly, direct nuclear radiation will be the dominant isomer-decay channel at $E_{i.s.}$ less than the difference $E_b(7P_{1/2}) - E_b^{(i)}$ between the binding energies of the excited atomic state $7P_{1/2}$ (about -2.9 eV according to our estimates) and the initial state [$E_b(6D_{3/2}) = -4.2$ eV in our calculation]. The reason is that an elastic electron bridge with $6D_{3/2}$ through an excited state $6D_{5/2}$ (and others) and with $7S_{1/2}$ through $6D_{3/2}$ (and other higher ones) has a relatively low probability. The half-life of an isomer in the region $E_{i.s.} \leqslant 1.3$ eV can be tens of days and more (at $F_{M1} = 10^{-2}$ and $E_{i.s.} = 1$ eV the lifetime relative to a radiative transition is approximately 27 days). Of course, if the energy ω_N is at resonance with the energy of one of the allowed atomic transitions, for example $6D_{3/2} \to 6D_{5/2}$ or $7S_{1/2} \to 6D_{3/2}$, an elastic electron bridge "works" very rapidly.

If $E_{i.s.}$ turns out to higher than the energy of the $7P_{1/2}$ level relative to the atomic ground state, then as $E_{i.s.}$ grows gradually the dominant decay channel is an inelastic bridge with population of the indicated level. In this case there will be initially emitted a γ quantum of energy $\omega_1 = E_{i.s.} - (E_b(7P_{1/2}) - E_b^{(i)})$, and then a photon (or photons, if population of higher states is energywise allowed) upon de-excitation of the atom. The spectrum of the possible isomer lifetimes is here very broad, from tens of hours to microseconds for resonance between the energies of the nuclear and one of the allowed quantum numbers of the atomic transitions.

Even though the atomic-state energies calculated by various programs can have a spread comparable with $E_{i.s.}$, the estimates given here are qualitatively valid. It is necessary to know more accurately the energy of the lowest of the $7P_{1/2,3/2}$ states, which "open" the channel of decay via an inelastic electron bridge. Since the levels indicated are low-lying, their energies are calculate with practically the same accuracy as the energies of the unexcited shells.

To conclude this section, we note the $E2$ transition admixture to the isomer nuclear $M1$ transition can be neglected in our case. In the long-wave approximation (our isomer-transition wavelength $\lambda \geqslant 2.5 \cdot 10^{-5}$ cm is appreciably larger than the radius R_N of the nucleus) the radiation probability per unit of the large multipole is $(R_N/\lambda)^2$ times larger, so that the value for the given transition is $\leqslant 10^{-15}$. According to the results of Ref. 4 the reduced probabilities of nuclear $E2$ transitions between the rotational bands to which the ground and isomer states belong are equal to several Weisskopf units. The $M1$ transition, as already mentioned, is attenuated by approximately two orders compared to electron wave functions $f(r)$, which enter in the radial matrix elements of the magnetic transitions, are of the order of $e^2 g(r)$. We obtain thus for the ratios of the $E2$ and $M1$ probabilities $(R_N/\lambda)^2 F_{E2}/F_{M1} e^4 \leqslant 10^{-8}$.

4. PRODUCTION OF ISOMER NUCLEI

According to presently available data[1] the probability of populating the first excited state of thorium-229 in α decay of uranium-233 directly and in γ transitions from higher states is very low—a fraction of a percent. It may therefore be more realistic to determine the characteristics of the investigated isomer level by an experimental setup in which enough excited nuclei are obtained in times $t \leqslant T_{1/2}$ by one or another method of pumping a nuclear transition. This can be, for example, a plasma with a temperature of several electron volts, produced by laser radiation or by an electronic beam. Successful experiments of this type are described in Refs. 15 and 16. From among the known mechanisms for stimulating nuclear transitions in a plasma [inelastic electron scattering, reverse internal electron conversion (RIEC), interaction of nuclei with the plasma's own thermal radiation, excitations in electron transitions and others (for details see Ref. 17)], the largest yield of isomer nuclei should be obtained in our case by the RIEC mechanism proposed in Ref. 18.

The processes named lead to formation of excited nuclei

only if the lifetimes of the excited atomic levels are shorter than the times of the conversion nuclear decay via these states. Otherwise, when the plasma cools down and the electrons begin to populate excited atomic levels, the already produced isomer nuclei decay rapidly. Calculations show that the characteristic times of conversion via excited atomic states, at $E_{i.s.} = 1$ to 5 eV, are not less than 10^{-5} s, so that deexcitation as a result of such a process has low probability.

The number N^* of isomer states produced in a plasma of temperature T and electron density n_e, during its lifetime τ, is given by

$$N^* = N n_e \tau \sigma v, \tag{5}$$

where N is the total number of ^{229}Th nuclei in the plasma volume, v is the electron velocity, σ the RIEC cross section given for a plasma with a Maxwellian electron-energy distribution by

$$\sigma = \frac{\pi}{m\varepsilon} 2\left(\frac{2m}{\pi T}\right)^{1/2} \exp(-\varepsilon/T)\frac{\Gamma_c}{T}, \tag{6}$$

Γ_c is the total conversion width of the nuclear level with allowance to decays via excited atomic states that are in thermodynamic equilibrium with the plasma and ε is the plasma electrons that excite resonantly the nuclei: $\varepsilon = E_{i.s.} + E_b$, where E_b is the binding energy of the populated atomic level. The largest width is from the states $8S_{1/2}$ and $9S_{1/2}$. With allowance for the attenuation coefficient assumed in Sec. 2 for the intensity of the nuclear $M1$ transition, a numerical calculation yields for Γ_c a value $\approx 10^{-12}$ eV in the region 1 eV $\leqslant E_{i.s.} \leqslant$ 5 eV. We obtain accordingly from Eq. (6) for the cross section the estimate $\approx 10^{-25}$ cm^2.

The effectiveness, defined as the ratio N^*/N in a plasma with a characteristic parameter value $n_e\tau \approx 10^{12}$ cm^{-3}·s, is approximately equal to 10^{-5}. Therefore if, for example, 10^{-6} g of ^{229}Th is used in the experiment, the yield is about 10^{10} isomer nuclei. The probability $W_\gamma^{(1)}$ of γ decay of the nucleus of an isomer-transition energy $\omega \approx 1$ eV is 10^{-6}–10^{-7} s^{-1}, i.e., we have even in this case 10^3–10^4 γ quanta per second.

If the decay is the result of IEC (see Sec. 2 above), the activity at millisecond isomer-state lifetimes is not less than 10^{13} Bk, whereas the α-decay activity does not exceed 10^5 Bk (^{229}Th is α-active with a half-life 7880 years[19]), and accordingly the induced α activity of the noise will also be significantly lower than the useful signal (by approximately two orders if it is assumed that the entire α-particle energy goes to formation of electrons with energies on the order of an electron volt).

5. CONCLUSION

An experimental determination of the energy and half-life of the first excited level in ^{229}Th with account taken of the estimates made here is quite complicated. The first step would therefore be a confirmation or denial of the existence of a conversion decay channel. Its presence would solve the problem completely, and its absence would yield an upper-bound estimate, independent of Ref. 2, of the nuclear-transition energy.

Decay via an electron bridge, as shown in Sec. 3, is possible in a rather large range of $E_{i.s.}$. It is seemingly possible, for the first time ever, to investigate this process in detail in "pure" form and not against the background of IEC that exceeds it by many orders.

A rather exotic phenomenon, nuclear emission in the optical band, will dominate for $E_{i.s.}$ smaller than approximately one and a half electron volts. More probable above this values, up to the ionization energy of the atom, is an EB. If one of these channels is actually realized, physicists will in time acquire a high-accuracy tool for the determination of subtle effects in atoms, in the structure of a solid, and others.

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