# Nuclear-spin mixing oscillations in $^{229}Th^{89+}$

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The $^{229}$Th nucleus is known to have a $3/2^+$ level at $3.5\pm1.0$ eV above the $5/2^+$ ground state. In a hydrogenlike $^{229}Th^{89+}$ ion these two levels are predicted to mix due to the high magnetic field produced at the nucleus by the 1*s* electron. This paper gives a theoretical analysis of the time evolution of the $^{229}Th^{89+}$ hydrogenlike system after its formation in a fast collision process. In particular, nuclear-spin mixing oscillations are considered and the oscillation frequency is estimated to be about $10^{15}$ s$^{-1}$. The time dependence of population of the hyperfine structure levels is determined. Considerations are extended to other ions of $^{229}$Th with the last electron in a higher *s* state. Consequences of different energy of the isomer are examined. A possible way to confront these predictions with experiment is briefly outlined.

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## I. INTRODUCTION

Precision $\gamma$-spectroscopy studies on the $\alpha$ decay of $^{233}$U led Helmer and Reich [1] to place the first excited state of the daughter $^{229}$Th at the unusually low energy of $3.5\pm1.0$ eV. This is the lowest excited state that has ever been found in an atomic nucleus. The spin and parity of this state was deduced as $3/2^+$, while that of the ground state is $5/2^+$ [1].

The half-life for the 3.5-eV $M1$ $\gamma$ transition in a fully stripped ion was estimated as 4 h [2]. In a neutral atom the decay can be faster due to an electronic bridge [3]. This provides competition to the $\gamma$ decay from a two-step process, in which part of the excitation energy is used for shifting a valence electron to a higher atomic orbit, while the rest is carried away by a "red-shifted" $\gamma$-ray photon. An enhancement of the isomeric transition due to this effect can be as high as three orders of magnitude [4].

An early search for the low-energy radiation accompanying the $^{233}U\rightarrow^{229}Th$ decay brought an observation of ultraviolet and visible photons [5,6]. However, this low-energy radiation has been shown to be caused by $\alpha$-particle induced fluorescence of air rather than by the deexcitation of the 3.5-eV isomer [7–9], so further experiments are needed.

A possibility to study the coupling of nuclear and atomic degrees of freedom is one of the reasons for particular interest in the low-energy isomer in $^{229}$Th [3,4,10–13]. The other reason is related to the nuclear-spin mixing that is predicted to take place in the highly charged $^{229}$Th ion. This mixing has already been theoretically studied in Refs. [2,14]. In the following section, results of these mixing studies are briefly reviewed. This is the ground for a theoretical analysis of a time evolution of the hydrogenlike $^{229}Th^{89+}$ ion after its formation in a relativistic collision process (e.g., via the fragmentation of a $^{238}$U beam or the in-flight stripping of electrons from a $^{229}$Th projectile). The emphasis is put on the nuclear-spin mixing oscillations that are expected to occur immediately after the ion is formed. To our knowledge, such oscillations have never been considered in nuclear physics. It is the near degeneracy, on the eV scale, of the two states in $^{229}$Th, which opens a possibility to observe oscillations. Our intention is to find to what extent these oscillations affect the time evolution of the hyperfine structure (hfs) levels.

We discuss briefly an experimental possibility to verify our theoretical predictions, to provide the first direct evidence for the existence of the isomer, and to measure more accurately its energy. Our discussion is extended to a possible use of the Li-like ($^{229}Th^{87+}$) and Na-like ($^{229}Th^{79+}$) ions. We also consider a possibility that energy of the isomer is outside the interval corresponding to the experimental uncertainty limits 2.5–4.5 eV.

## II. NUCLEAR-SPIN MIXING IN $^{229}Th^{89+}$

We consider $^{229}$Th with all but one electron removed. The remaining electron stays in the $K$ shell. Its intrinsic magnetic moment produces a magnetic field of about 28 MT at the nucleus [2]. Due to the hyperfine interaction, the $5/2^+$ ground state is split into $F=2$ (lower) and $F=3$ components, while the $3/2^+$ isomer is split into $F=2$ (upper) and $F=1$ components. These components are labeled $|1\rangle$ through $|4\rangle$ according to the increasing energy, as shown in Fig. 1. Instead of one isomeric $M1$ transition, now five such transitions are possible. The latter are characterized by widths $\gamma_{fi}$.

The magnetic field of the electron leads also to a mixing of the two $F=2$ states. This nuclear-spin mixing can be expressed in the following way:

$$\psi_3=\cos(\theta)\psi[3/2]+\sin(\theta)\psi[5/2],$$

$$\psi_1=-\sin(\theta)\psi[3/2]+\cos(\theta)\psi[5/2]. \quad (1)$$

Here $\psi[5/2]$ and $\psi[3/2]$ denote the $F=2$ hydrogenlike-

 

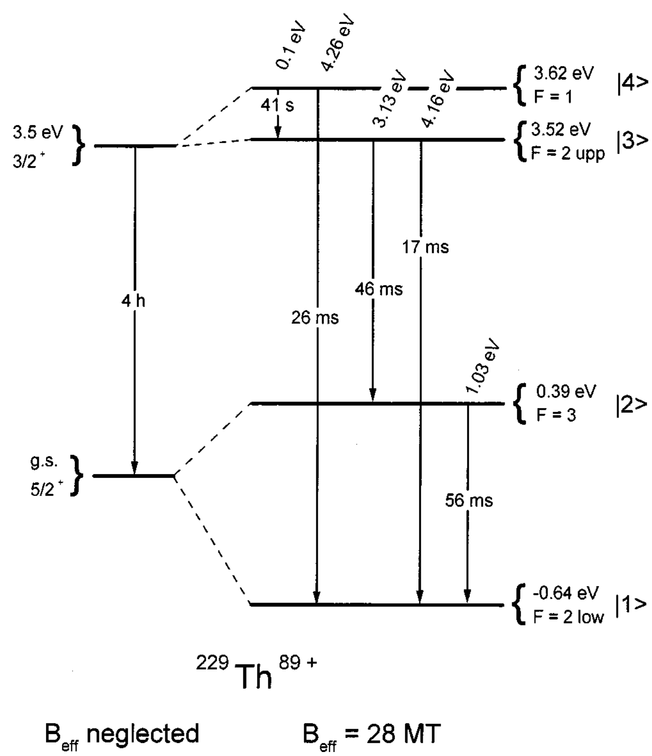


FIG. 1. Predictions for the hydrogenic ion of $^{229}$Th with an electron in the $K$ shell. Left: An abstract situation with the magnetic interaction "switched off." The energy distance between the $5/2^+$ ground and $3/2^+$ isomeric nuclear states [1] is not perturbed by the hyperfine interaction. The estimate of the nuclear $M1$ $\gamma$-transition half-life is taken from Ref. [2]. Right: The hfs levels are shown with energies given (not to scale) relative to the unperturbed ground state [2]. Due to the nuclear-spin mixing, transitions between these levels are the $M1$ electron spin-flip transitions. For each transition, the calculated energy and mean lifetime are taken from [2] and [14], respectively. The relevant widths, used in the text, are $\gamma_{fi}=1/\tau(i\rightarrow f)$.

ion states with the nucleus in the $I=5/2$ and 3/2 states, respectively. The relation between the mixing parameters $a$ and $b$ of Ref. [2], the parameter $\beta$ of Ref. [14], and the parameter $\theta$ used here is given by

$$a=1/\sqrt{1+\beta^2}=\cos(\theta) \quad \text{and} \quad b=\beta/\sqrt{1+\beta^2}=\sin(\theta).$$

The mixing parameters depend on the isomer energy and for 3.5 eV one gets $\sin^2(\theta)=0.02$ [2]. Due to the mixing effect, the radiative transitions between the upper and lower doublets of the hfs levels are essentially electron spin-flip transitions [14]. Compared to the nuclear $\gamma$-ray transition these are enhanced by several orders of magnitude.

The spin admixtures considered above correspond to the stationary conditions. However, one may ask the question what happens when the magnetic field is "turned on" at $t=0$ and the ion evolves in time. In the first step, discussed in Sec. III, we neglect the radiative transitions. Then the wave functions are

$$\psi_k(t)=\psi_k \exp(-iE_k t), \tag{2}$$

where $k=1$ to 4 (we use natural units $\hbar=c=1$). Our attention is focused on the $F=2$ components, that is on the states $|1\rangle$ and $|3\rangle$.

In the next step, discussed in Sec. IV, we consider all four states. The state $|1\rangle$ is taken again as a stationary one since its $\alpha$-decay width can obviously be neglected. For the other three states we take into account the widths related to the $M1$ transitions shown in Fig. 1. The wave function of state $|4\rangle$ is simply

$$\psi_4(t)=\psi_4 \exp[-(iE_4+\Gamma_4/2)t],$$

where $\Gamma_4=\gamma_{14}+\gamma_{34}$. However, states $|2\rangle$ and $|3\rangle$ are not only deexcited but also fed by $M1$ transitions. Hence, the density-matrix formalism [15] is applied to find the time evolution of this system.

## III. SPIN-MIXING OSCILLATIONS IN THE $F=2$ STATES, DECAY NEGLECTED

We assume that the hydrogenlike ion $^{229}$Th$^{89+}$ is formed, and the magnetic field is "turned on," at $t=0$. Now, we are interested in the propagation of this system. In general, it is described by a wave function $\Phi(t)$ that is a superposition of the basic states $|1\rangle$ through $|4\rangle$. However, in this section we focus our attention on the $F=2$ states and disregard the population of the $F=1,3$ states. With the decay process neglected ($\gamma_{fi}=0$), our wave function becomes a superposition of two basic states

$$\Phi(t)=a_1\psi_1(t)+a_3\psi_3(t). \tag{3}$$

The coefficients $a_1$ and $a_3$ are determined by the initial conditions. To be specific, we begin with the assumption that all nuclei are originally (for $t=0$) in the $I=3/2$ isomeric state. It is also assumed that the process of creation of the $^{229}$Th$^{89+}$ ion is very fast, allowing for a sudden approximation (the time of flight of a fast projectile over the diameter of the $K$-electron orbit gives an estimated interaction time of $10^{-20}$ s). With this initial condition, Eq. (1) generates the time dependence of the system

$$\Phi_m(t)=-\sin(\theta)\psi_1(t)+\cos(\theta)\psi_3(t), \tag{4}$$

where the index "$m$" is added to indicate the isomeric state at $t=0$. This expression assures $\Phi_m(0)=\psi[3/2]$. The probability $P_m[F,I;t]$ to find the nucleus in the spin $I=3/2$ state at some later time $t$ is given by

$$\begin{aligned} P_m[2,3/2;t]&=|\langle\Phi_m(t)|\psi[3/2]\rangle|^2\\ &=1-\sin^2(2\theta)\sin^2(\Omega_{13}t/2), \end{aligned} \tag{5}$$

where $\Omega_{13}=E_3-E_1$. Obviously, the probability to find the same $F=2$ ion state with the nucleus having spin 5/2 is

$$P_m[2,5/2;t]=1-P_m[2,3/2;t]=\sin^2(2\theta)\sin^2(\Omega_{13}t/2). \tag{6}$$

For the isomer energy of 3.5 eV, the difference between the energies of the two $F=2$ states is 4.16 eV, see Fig. 1. In natural units it corresponds to $\Omega_{13} = 6.3\times10^{15}\ \mathrm{s}^{-1}$. The relevant period of oscillations is $t_{osc} = 2\pi/\Omega_{13} \approx 10^{-15}$ s.

The oscillations considered here can indirectly manifest themselves in another way. For $\sin^2(\theta)\approx 0.02$, the time averaged probability of finding the $I=5/2$ component, when the initial state is $\psi[3/2]$, becomes

$$\overline{P_m[2,5/2;t]} = (1/2)\sin^2(2\theta) \approx 0.04.$$

With the oscillations present, the probability of finding the $I=5/2$ component is about twice as large as that of finding the same component in the stationary $F=2$ (upper) state $\psi_3$, see Sec. II. To explain this difference in more detail one has to account for the instability of the hfs levels involved. This is done in the following section.

Other initial conditions may also be considered. One can start with the $^{229}Th$ nuclei in the $I=5/2$ ground state. With the creation of the hydrogenlike ion at $t=0$, there will be oscillations of the probability to find the spin 3/2 admixture

$$P_g[2,3/2;t] = 1 - P_g[2,5/2;t] = \sin^2(2\theta)\sin^2(\Omega_{13}t/2) \tag{7}$$

and the time averaged value $\overline{P_g[2,3/2;t]}$ will be about 4%.

## IV. OSCILLATIONS WITH DECAY INCLUDED

We assume again formation of the hydrogenlike ion $^{229}Th^{89+}$ at $t=0$. Now, the radiative transitions are accounted for.

### A. Nuclei originally in the isomeric state

It is assumed that at $t=0$ all nuclei are in the $I=3/2$ isomeric state. The task is to calculate the probability of finding the hydrogenlike ion with the nucleus in the $I=3/2$ state for $t>0$. This probability is a sum of terms corresponding to the $F=2$ and the unmixed $F=1$ ion states,

$$P_m[3/2;t] = P_m[2,3/2;t] + P_m[1,3/2;t]. \tag{8}$$

The index "$m$" is added, as before, to indicate the isomeric state at $t=0$. To find the $P_m[F,I;t]$ terms, the density-matrix approach is applied [15]. It is assumed that the population of the two hfs states at $t=0$ is proportional to $2F+1$.

With reference to Sec. III, the $F=2$ state of the ion at $t=0$ is given by

$$\Phi_m(0) = -\sin(\theta)\psi_1 + 0\psi_2 + \cos(\theta)\psi_3 + 0\psi_4. \tag{9}$$

The relevant density matrix, which accounts for the statistical population of the $F=2$ and $F=1$ states, is

$$\varrho_m(0) = |\Phi_m(0)\rangle \frac{5}{8}\langle\Phi_m(0)| + |4\rangle\frac{3}{8}\langle 4|$$
$$= \begin{bmatrix} \frac{5}{8}\sin^2(\theta) & 0 & -\frac{5}{8}\sin(\theta)\cos(\theta) & 0 \\ 0 & 0 & 0 & 0 \\ -\frac{5}{8}\sin(\theta)\cos(\theta) & 0 & \frac{5}{8}\cos^2(\theta) & 0 \\ 0 & 0 & 0 & \frac{3}{8} \end{bmatrix}. \tag{10}$$

The rows and columns of this matrix correspond to the four basic states of Fig. 1. The time dependence of the density matrix $\varrho_m(t)$ can be found from equations given in the Appendix together with their solutions. These equations account for four transitions only. The $|4\rangle\to|3\rangle$ transition is neglected as it is relatively a very slow one. With the $\varrho_m(t)$ available one has

$$P_m[2,3/2;t] = \langle\Phi_m(0)|\varrho_m(t)|\Phi_m(0)\rangle. \tag{11}$$

The final expression for this probability reads

$$\begin{aligned} P_m[2,3/2;t] = \frac{5}{8}\Bigg\{ & \cos^4(\theta)\exp[-(\gamma_{13}+\gamma_{23})t] + \sin^4(\theta) \\ & + \sin^2(\theta)\cos^2(\theta)\Bigg\{1 + \exp[-(\gamma_{13}+\gamma_{23})t] \\ & \times \frac{\gamma_{13}-\gamma_{12}}{\gamma_{12}-\gamma_{13}-\gamma_{23}} \\ & + \exp(-\gamma_{12}t)\frac{\gamma_{23}}{\gamma_{12}-\gamma_{13}-\gamma_{23}}\Bigg\} \\ & + \frac{3}{5}\sin^2(\theta)[1-\exp(-\gamma_{14}t)] \\ & + [2\cos^2(\theta)\sin^2(\theta) - \sin^2(2\theta)\sin^2(\Omega_{13}t/2)] \\ & \times \exp[-(\gamma_{13}+\gamma_{23})t/2]\Bigg\}. \end{aligned} \tag{12}$$

This expression accounts for those hydrogenlike ions that at $t=0$ were in the $F=2$ state. In the limit of very short times $t$ it reproduces expression (5), provided the latter is multiplied by 5/8.

An analogous procedure leads to a simple expression for the second term

$$P_m[1,3/2;t] = (3/8)\exp(-\gamma_{14}t). \tag{13}$$

It accounts for the decay of those ions that at $t=0$ were in the $F=1$ state, and in contrast to Eq. (12), agrees with simple intuition.

After averaging over time intervals equal to $t_{osc}$ [equivalent to removing the last term of Eq. (12)] both probabilities

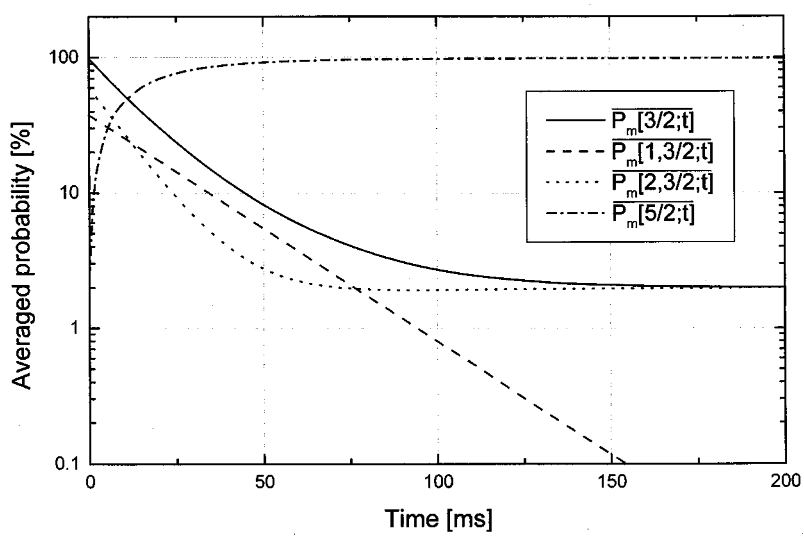


FIG. 2. Time dependence of probabilities $\overline{P_m[3/2;t]}$, $\overline{P_m[1,3/2;t]}$, $\overline{P_m[2,3/2;t]}$, and $\overline{P_m[5/2;t]}$, averaged over $t_{osc}$, to find the $^{229}\mathrm{Th}^{89+}$ ion in the state: $I=3/2$ ($F=2$ or $F=1$), $I=3/2$ ($F=1$), $I=3/2$ ($F=2$), and $I=5/2$ ($F=2$ or $F=3$), respectively. It has been assumed that (i) the nucleus at $t\leqslant 0$ is in the pure $I=3/2$ isomeric state, (ii) the $^{229}\mathrm{Th}^{89+}$ ion is created at $t=0$, (iii) the mixing amplitude is $\sin(\theta)=\sqrt{0.02}$. At $t=0$, these probabilities are 97.5, 37.5, 60, and 2.5 %, respectively. At $t\to\infty$, these are 2, 0, 2, and 98 %.

are shown in Fig. 2 together with their sum $\overline{P_m[3/2;t]}=\overline{P_m[2,3/2;t]}+\overline{P_m[1,3/2;t]}$ and with $\overline{P_m[5/2;t]}$. For $t=0$ one finds $P_m[2,3/2;0]=0.6$ and $P_m[2,5/2;0]=0.025$ (as expected, see Sec. III, the ratio of these probabilities is 96/4).

### B. Nuclei originally in the ground state

Now, for $t=0$ we assume that all nuclei are in the $I=5/2$ state. The probability of finding the hydrogenlike ion with the nucleus in the $I=5/2$ state at $t>0$ is taken as a sum of terms corresponding to the $F=2$ and $F=3$ ion states,

$$P_g[5/2;t]=P_g[2,5/2;t]+P_g[3,5/2;t]. \tag{14}$$

The index "$g$" indicates that the nuclei at $t=0$ are in the ground state. To find $P_g[F,I;t]$ terms, again the density-matrix approach is applied and the population of the two hfs states at $t=0$ is assumed to be proportional to $2F+1$. The state $|4\rangle$ plays no role here, and the $F=2$ state of the ion at $t=0$ is given by

$$\Phi_g(0)=\cos(\theta)\psi_1+0\,\psi_2+\sin(\theta)\psi_3. \tag{15}$$

The relevant density matrix is now a $3\times 3$ matrix

$$\varrho_g(0)=|\Phi_g(0)\rangle\frac{5}{12}\langle\Phi_g(0)|+|2\rangle\frac{7}{12}\langle 2| = \begin{bmatrix} \frac{5}{12}\cos^2(\theta) & 0 & \frac{5}{12}\sin(\theta)\cos(\theta) \\ 0 & \frac{7}{12} & 0 \\ \frac{5}{12}\sin(\theta)\cos(\theta) & 0 & \frac{5}{12}\sin^2(\theta) \end{bmatrix}. \tag{16}$$

By finding the time dependence of the density matrix, $\varrho_g(t)$, in the way analogous to that described in Sec. IV A and in the Appendix, one gets the probability to find the $F=2$ hydrogenlike-ion state with the nucleus having spin 5/2 (under the assumption that at $t=0$ all nuclei were in the $I=5/2$ ground state):

$$\begin{aligned} P_g[2,5/2;t]=\frac{5}{12}\Bigg\{&\sin^4(\theta)\exp[-(\gamma_{13}+\gamma_{23})t]+\cos^4(\theta) \\ &+\sin^2(\theta)\cos^2(\theta)\Bigg\{1+\exp[-(\gamma_{13}+\gamma_{23})t] \\ &\times\frac{\gamma_{13}-\gamma_{12}}{\gamma_{12}-\gamma_{13}-\gamma_{23}} \\ &+\exp(-\gamma_{12}t)\frac{\gamma_{23}}{\gamma_{12}-\gamma_{13}-\gamma_{23}}\Bigg\} \\ &+\frac{7}{5}\cos^2(\theta)[1-\exp(-\gamma_{12}t)] \\ &+[2\cos^2(\theta)\sin^2(\theta)-\sin^2(2\,\theta)\sin^2(\Omega_{13}t/2)] \\ &\times\exp[-(\gamma_{13}+\gamma_{23})t/2]\Bigg\}. \end{aligned} \tag{17}$$

A partial probability to find the upper $F=2$ state is given as

$$P_g[2_{\mathrm{upp}},5/2;t]=\langle 3|\varrho(t)|3\rangle=\frac{5}{12}\sin^2(\theta)\exp[-(\gamma_{13}+\gamma_{23})t]. \tag{18}$$

For the term corresponding to $F=3$ one obtains

$$P_g[3,5/2;t]=\left(\frac{7}{12}+\frac{\gamma_{23}}{\gamma_{12}-\gamma_{13}-\gamma_{23}}\{\exp[(\gamma_{12}-\gamma_{13}-\gamma_{23})t]-1\}\frac{5}{12}\sin^2(\theta)\right)\exp(-\gamma_{12}t). \tag{19}$$

The results of calculations, averaged over time as in the previous case, are shown in Fig. 3.

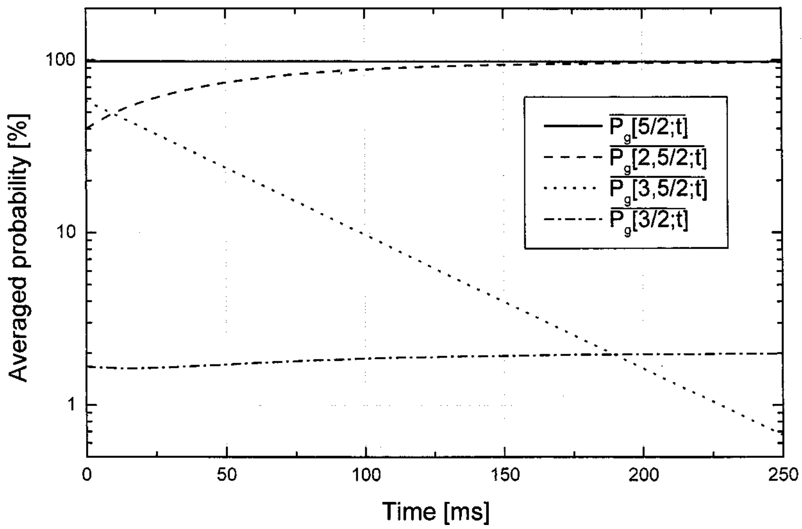


FIG. 3. Time dependence of probabilities $\overline{P_g[5/2;t]}$, $\overline{P_g[2,5/2;t]}$, $\overline{P_g[3,5/2;t]}$, and $\overline{P_g[3/2;t]}$, averaged over $t_{osc}$, to find the $^{229}Th^{89+}$ ion in the state: $I=5/2$ ($F=2$ or $F=3$), $I=5/2$ ($F=2$), $I=5/2$ ($F=3$), and $I=3/2$ ($F=1$ or $F=2$), respectively. It has been assumed that the nucleus at $t\leqslant 0$ is in the pure $I=5/2$ ground state and the conditions (ii) and (iii), specified in the caption to Fig. 2, are also fulfilled. At $t=0$, these probabilities are 98.3, 40, 58.3, and 1.7 %, respectively. At $t\rightarrow\infty$, these are 98, 98, 0, and 2 %.

## V. TOWARDS EXPERIMENTS

To illustrate a possible way to verify the predictions of this paper, we consider two experiments that would be difficult at present but may become realistic in the future. We assume that (i) the $^{229}Th^{89+}$ hydrogenlike ions, produced at $t=0$, are circulating in a storage ring, and (ii) the photons originating from deexcitation of the higher hfs levels can be detected with a reasonable efficiency and resolution. For the sake of simplicity we disregard here the 1-eV uncertainty of the isomer energy (and the uncertainties of other nuclear parameters that determine the nuclear-spin mixing: nuclear magnetic moments of the isomer and of the ground state, as well as the reduced probability of the $M1$ transition between these states, see discussion in [2,14]).

First, referring to Sec. IV B, we assume that all $^{229}Th$ nuclei at $t=0$ are in the $5/2^+$ ground state. Directly after formation of $^{229}Th^{89+}$, this hydrogenlike system is expected to be in three hfs states. In addition to the $F=3$ and the lower $F=2$ levels, the upper $F=2$ level should also be populated due to the nuclear-spin-mixing mechanism. In the photon spectrum, the 1.03-eV line should be accompanied by two weaker lines at 3.13 and 4.16 eV, see Fig. 1. Observation of the latter two lines would be the first direct evidence for the existence of the isomeric state. It would also allow for a better determination of the isomer energy. Such an experiment, combined with the laser experiment considered in Ref. [14], would improve the nuclear data essential for our predictions.

As follows from Eq. (19), the intensity of the 1.03-eV transition is proportional to $\gamma_{12}P_g[3,5/2;t] \approx (7/12)\gamma_{12}\exp(-\gamma_{12}t)$, while a joint intensity of the two other transitions is proportional to $(5/12)(\gamma_{13}+\gamma_{23})\sin^2(\theta)\exp[-(\gamma_{13}+\gamma_{23})t]$. The ratio of these intensities is given by

$$R\approx\frac{5(\gamma_{13}+\gamma_{23})}{7\gamma_{12}}\sin^2(\theta)\exp[(\gamma_{12}-\gamma_{13}-\gamma_{23})t]. \quad (20)$$

A comparison of an experimental intensity ratio with $R$ would be a partial test for our predictions. Unfortunately, it would be insensitive to the nuclear-spin-mixing oscillations.

In view of the very high frequency involved, a direct observation of the oscillations is rather impossible. An indirect evidence for this effect could be obtained in the following experiment. Assume that originally the $^{229}Th^{88+}$ heliumlike ions are circulating in a storage ring, and all nuclei are in the $5/2^+$ ground state. No magnetic interaction and no nuclear-spin mixing occurs. By stripping off one electron at $t=0$ these ions are converted into the $^{229}Th^{89+}$ systems. The magnetic field and the nuclear-spin-mixing oscillations are "turned on." Next, at $t=T$ the second electron is stripped off and the effect is removed. Afterwards, some of the nuclei remain in the isomeric $I=3/2$ state. A measurement of the population of the isomer is to be performed. If the time interval $T$ is chosen to be much shorter than the mean lifetimes, the isomer population should be 1.7%, see Fig. 3. This value reflects the enhancement of the nuclear-spin mixing due to the effect of oscillations. Without the oscillations, it would be lower by a factor of about 2, as shown in Sec. III.

## VI. SENSITIVITY TO THE ION CHARGE AND TO THE ISOMER ENERGY

### A. Use of other ions

As in the preceding sections, we assume here the isomer energy to be exactly 3.5 eV. A question is what happens if $^{229}Th^{89+}$ is replaced by another ion having an odd number of electrons. We mean an ion with the last electron occupying a higher ns state and yielding an effective magnetic field at the nucleus of about $28/n^3$ MT.

For the $^{229}Th^{87+}$ Li-like ion, $n=2$ and $B_{eff}\approx 3.5$ MT. Then, the energies of the hfs levels 1 through 4 (see Fig. 1) are $-0.070$, 0.049, 3.493, and 3.515 eV, respectively, and the spin-mixing probability, $\sin^2\theta$ is equal to 0.043%. The mean lifetimes of the $|4\rangle\rightarrow|3\rangle$, $|4\rangle\rightarrow|1\rangle$, $|3\rangle\rightarrow|2\rangle$, $|3\rangle\rightarrow|1\rangle$, and $|2\rangle\rightarrow|1\rangle$ transitions are $4\times10^3$ s, 2.0 s, 1.6 s, 1.2 s, and 36 s, respectively. The spin-mixing oscillation period becomes $1.2\times10^{-15}$ s.

TABLE I. Energies of the hfs levels and mean lifetimes of the $M1$ transitions to the $F=2$ (low) state and to the $F=3$ state (in brackets) for $E_{isom}=10$ eV, and for three charge states of the $^{229}$Th ion.

| Ions | | H-like | | Li-like | | Na-like | |
|---|---|---|---|---|---|---|---|
| State | $F$ | $E_F$ (eV) | $\tau$ (s) | $E_F$ (eV) | $\tau$ (s) | $E_F$ (eV) | $\tau$ (s) |
| $\vert 4\rangle$ | 1 | 10.12 | 0.010 | 10.02 | 0.72 | 10.004 | 8.3 |
| $\vert 3\rangle$ | 2upp | 9.96 | 0.007 | 9.99 | 0.44 | 9.997 | 5.0 |
| | | | (0.010) | | (0.54) | | (6.0) |
| $\vert 2\rangle$ | 3 | 0.39 | 0.065 | 0.05 | 36.5 | 0.015 | $1.4\times10^3$ |
| $\vert 1\rangle$ | 2low | $-0.58$ | | $-0.07$ | | $-0.020$ | |

Almost the same oscillation period is obtained for the Na-like ion $^{229}$Th$^{79+}$ for which $n=3$ and $B_{eff}\approx1$ MT. The level energies are $-0.021$, 0.015, 3.498, and 3.504 eV. The mixing probability is only 0.004%, and the mean lifetimes of the transitions listed above are $1.3\times10^5$, 24, 18, 14, and $1.4\times10^3$ s, respectively.

For a description of the time evolution of these ions, the formulas given in Sec. IV are applicable directly. Although experimental predictions discussed in the previous section remain valid for the two cases considered here, the effects are much smaller due to the lower $B_{eff}$ value.

### B. Assumption of the isomer at 10 eV

The energy of the isomer, $3.5\pm1.0$ eV, was obtained [1] from differences of much higher energies of $\gamma$ rays. Possibly, the shift in energy exceeding 1 eV cannot be ruled out. It is worthwhile to examine the consequences of the energy being far outside the 2.5–4.5 eV interval.

First, we consider the isomer energy equal to 10 eV. The half-life of this state in a bare nucleus is close to 600 s. For the $^{229}$Th$^{89+}$, $^{229}$Th$^{87+}$, and $^{229}$Th$^{79+}$ ions, the mixing probability is 0.32%, 0.0054%, and 0.00048%, respectively. The period of nuclear-spin-mixing oscillations is approximately $4\times10^{-16}$ in each case. The order of the hfs levels is the same as in Fig. 1. The calculated energies of these levels are given in Table 1. This table includes also the mean lifetimes of those $M1$ transitions that are needed to study the time evolution of the system with the use of the formulas given in Sec. IV. The $|4\rangle\rightarrow|3\rangle$ transitions are relatively very slow and are disregarded.

The mixing effect is much weaker than that expected for $E_{isom}=3.5$ eV. Although the $M1$ transitions are still clearly enhanced relative to the $\gamma$ transition, detection of this effect might be difficult.

### C. Assumption of the complete degeneracy

To illustrate an opposite situation, we assume a complete degeneracy of the $5/2^+$ and $3/2^+$ nuclear states. The calculated energies of the hfs levels are given in Table II. The order of the three upper levels is changed compared to that of Fig. 1.

In Table II, the mean lifetimes are given also for the main $M1$ transitions. These lifetimes are practically equal to the mean lifetimes of the levels because the omitted transitions are much slower. Therefore, one may assume that each of these levels decays exponentially.

For each of the three ions, the nuclear-spin-mixing probability is 31%. With the decreasing charge of the ion the oscillation period increases from $3.2\times10^{-15}$, through $2.6\times10^{-14}$ to $8.8\times10^{-14}$ s. If we assume that the nucleus at $t\leqslant0$ is in the pure $I=5/2$ ground state, the level-population probabilities at $t=0$ are found to be $\overline{P[5/2;t]}=82\%$, $\overline{P[2,5/2;t]}=24\%$, $\overline{P[3,5/2;t]}=58\%$, and $\overline{P[3/2;t]}=18\%$. As earlier, these are the probabilities averaged over the oscillation period, see Sec. IV and Fig. 3.

For an experimentalist, the case of $E_{isom}=0$ could be attractive. The spin-mixing effect is large. The role of the mixing oscillations is pronounced. However, although the period of oscillations is much longer than for $E_{isom}=3.5$ eV, their direct observation will be still nearly impossible. In addition, rather low energies of the transitions may be inconvenient for detection purposes.

## VII. SUMMARY

The doublet of the $3/2^+$ isomeric and $5/2^+$ ground states of the $^{229}$Th nucleus offers a unique chance to study the coupling of nuclear and atomic degrees of freedom. However, for an effective use of this chance, the isomer energy

TABLE II. Energies of the hfs levels and mean lifetimes of the $M1$ transitions to the $F=2$ (low) state for $E_{isom}=0$, and for three charge states of the $^{229}$Th ion.

| Ions | H-like | | Li-like | | Na-like | |
|---|---|---|---|---|---|---|
| $F$ | $E_F$ (eV) | $\tau$ (s) | $E_F$ (eV) | $\tau$ (s) | $E_F$ (eV) | $\tau$ (s) |
| 3 | 0.393 | 0.036 | 0.049 | 19 | 0.015 | 715 |
| 2upp | 0.331 | 0.054 | 0.041 | 28 | 0.012 | 1060 |
| 1 | 0.116 | 0.11 | 0.015 | 55 | 0.004 | 2090 |
| 2low | $-0.951$ | | $-0.119$ | | $-0.035$ | |

has to be determined with a better accuracy. Hopefully, the latter can be achieved via studies of the nuclear-spin-mixing effect. This effect is expected to take place in $^{229}Th^{89+}$ due to the magnetic interaction between the $1s$ electron and the nucleus. It should manifest itself in the splitting of the hfs levels, and in the enhancement of the $M1$ transitions between these levels relative to the $3/2^+ \to 5/2^+$ nuclear transition.

This work gives the theoretical analysis of the time evolution of the $^{229}Th^{89+}$ system after its formation in a fast collision process. Expressions were presented for the time dependence of the population of the hfs levels, and for the intensities of the $M1$ transitions in particular case of the pure $5/2^+$ nuclear state at $t=0$. The calculated transition intensities can be directly compared with results of a future decay experiment. A better determination of the nuclear parameters would be one of the ultimate goals of the experimental studies.

In the theoretical considerations, the emphasis was put on clarification of the role of the nuclear-spin-mixing oscillations. We have shown that the oscillation frequency is as high as about $10^{15}$ Hz. Therefore, these oscillations are rather difficult for a direct observation. However, an indirect experimental evidence could be, perhaps, obtained.

There may be practical arguments in favor of using Li-like and Na-like ions in the experiment instead of H-like ions. Therefore, the considerations were extended to include these two cases. It was found, however, that the predicted effects are much weaker than in the case of the H-like system that remains to be the most promising one for experimental studies. Additionally, consequences of the isomer energy being different from the literature value were briefly discussed.

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## APPENDIX

With reference to Sec. IV A, the set of master equations for the density-matrix elements (with the index "$m$" omitted) is

(i) $$\frac{d}{dt}\varrho_{11}=\gamma_{14}\varrho_{44}+\gamma_{13}\varrho_{33}+\gamma_{12}\varrho_{22},$$

(ii) $$\frac{d}{dt}\varrho_{22}=\gamma_{23}\varrho_{33}-\gamma_{12}\varrho_{22},$$

(iii) $$\frac{d}{dt}\varrho_{33}=-(\gamma_{13}+\gamma_{23})\varrho_{33},$$

(iv) $$\frac{d}{dt}\varrho_{44}=-\gamma_{14}\varrho_{44},$$

(v) $$\frac{d}{dt}\varrho_{12}=-i\Omega_{12}\varrho_{12}-\frac{1}{2}\gamma_{12}\varrho_{12},$$

(vi) $$\frac{d}{dt}\varrho_{13}=-i\Omega_{13}\varrho_{13}-\frac{1}{2}(\gamma_{13}+\gamma_{23})\varrho_{13},$$

(vii) $$\frac{d}{dt}\varrho_{23}=-i\Omega_{23}\varrho_{23}-\frac{1}{2}(\gamma_{23}+\gamma_{13}+\gamma_{12})\varrho_{23},$$

(vii) $$\frac{d}{dt}\varrho_{14}=-i\Omega_{14}\varrho_{14}-\frac{1}{2}\gamma_{14}\varrho_{14}.$$

We have neglected here the $|4\rangle\to|3\rangle$ transition as a relatively very slow one. The solutions for individual matrix elements are

$$\varrho_{14}(t)=\varrho_{14}(0)\exp(-i\Omega_{14}t-\gamma_{14}t/2)=\varrho^*_{41}(t),$$

$$\varrho_{23}(t)=\varrho_{23}(0)\exp\left[-i\Omega_{23}t-\frac{t}{2}(\gamma_{23}+\gamma_{13}+\gamma_{12})\right]=\varrho^*_{32}(t),$$

$$\varrho_{13}(t)=\varrho_{13}(0)\exp\left[-i\Omega_{13}t-\frac{t}{2}(\gamma_{13}+\gamma_{23})\right]=\varrho^*_{31}(t),$$

$$\varrho_{12}(t)=\varrho_{12}(0)\exp\left(-i\Omega_{12}t-\frac{t}{2}\gamma_{12}\right)=\varrho^*_{21}(t),$$

$$\varrho_{44}(t)=\varrho_{44}(0)\exp(-\gamma_{14}t),$$

$$\varrho_{33}(t)=\varrho_{33}(0)\exp[-(\gamma_{13}+\gamma_{23})t],$$

$$\varrho_{22}(t)=\left\{\varrho_{22}(0)+\frac{\gamma_{23}}{\gamma_{12}-\gamma_{13}-\gamma_{23}}\varrho_{33}(0)[\exp(\gamma_{12}-\gamma_{13}-\gamma_{23})t-1]\right\}\exp(-\gamma_{12}t),$$

$$\begin{aligned}\varrho_{11}(t)=\varrho_{11}(0)+\varrho_{22}(0)[1-\exp(-\gamma_{12}t)]\\ &+\varrho_{33}(0)\left\{1+\exp[-(\gamma_{13}+\gamma_{23})t]\frac{\gamma_{13}-\gamma_{12}}{\gamma_{12}-\gamma_{13}-\gamma_{23}}\right.\\ &\left.+\exp(-\gamma_{12}t)\frac{\gamma_{23}}{\gamma_{12}-\gamma_{13}-\gamma_{23}}\right\}\\ &+\varrho_{44}(0)[1-\exp(-\gamma_{14}t)].\end{aligned}$$

As expected, $\varrho_{ij}(t)\to\varrho_{ij}(0)$ for $t\to 0$.

[1] R.G. Helmer and C.W. Reich, Phys. Rev. C **49**, 1845 (1994), and earlier papers quoted therein.
[2] S. Wycech and J. Żylicz, Acta Phys. Pol. B **24**, 637 (1993).
[3] E.V. Tkalya, Pisma Zh. Éksp. Teor. Fiz. **55**, 216 (1992) [JETP Lett. **55**, 211 (1992)].
[4] F.F. Karpeshin, I.M. Band, M.B. Trzhaskovskaya, and A. Pastor, Phys. Rev. Lett. **83**, 1072 (1999).
[5] G.M. Irwin and K.H. Kim, Phys. Rev. Lett. **79**, 990 (1997).
[6] D.S. Richardson, D.M. Benton, D.E. Evans, J.A.R. Griffith, and G. Tungate, Phys. Rev. Lett. **80**, 3206 (1998).
[7] S.B. Utter, P. Beiersdorfer, A. Barnes, R.W. Lougheed, J.R. Crespo Lopez-Urrutia, J.A. Becker, and M.S. Weiss, Phys. Rev. Lett. **82**, 505 (1999).
[8] R.W. Shaw, J.P. Young, S.P. Cooper, and O.F. Webb, Phys. Rev. Lett. **82**, 1109 (1999).
[9] J.P. Young, R.W. Shaw, and O.F. Webb, Inorg. Chem. **38**, 5112 (1999).
[10] F.F. Karpeshin, I.M. Band, M.B. Trzhaskovskaya, and B.A. Zonn, Phys. Lett. B **282**, 267 (1992).
[11] E.V. Tkalya, A.N. Zherikin, and V.I. Zhudov, Phys. Rev. C **61**, 064308 (2000).
[12] S. Matinyan, Phys. Rep. **298**, 199 (1998).
[13] F.F. Karpeshin, I.M. Band, and M.B. Trzhaskovskaya, Nucl. Phys. **A654**, 579 (1999).
[14] F.F. Karpeshin, S. Wycech, I.M. Band, M.B. Trzhaskovskaya, M. Pfützner, and J. Żylicz, Phys. Rev. C **57**, 3085 (1998).
[15] P. Meystre and M. Sargent, *Elements of Quantum Optics* (Springer, Berlin 1991).