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Induced decay of $^{178}\mathrm{Hf}^{m2}$: Theoretical analysis of experimental results

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This article reviews experimental results obtained recently on the x-ray-induced acceleration of the decay of the long-lived isomer $^{178}\mathrm{Hf}^{m2}$. Two basic mechanisms for the induced decay are considered: (1) direct interaction of the incident x rays with the nucleus and (2) the nucleus-x-ray interaction proceeding via atomic shells. We establish that the absence of K forbiddenness for all transitions to a hypothetical "mixed K" level cannot explain the measured cross sections even if collective nuclear matrix elements, resonant conditions, and so on, are assumed. We also tested, and rejected, the hypothesis that the enhancement is due to normal nuclear transitions in the inverse nuclear excitation by electron transition process. The possibility to make measurements with intense laser radiation is considered too. Thus, there appears to be no explanation of these experimental results within quantum electrodynamics and the contemporary concepts of atomic nuclei.

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I. INTRODUCTION

Controversial results on the x-ray-induced decay of the $^{178}\mathrm{Hf}^{m2}$ isomer (16^+, 2.446 MeV, 31 yr) have been published recently in a number of scientific journals by the research group of Collins [1–7]. Considerable interest in these experimental data is driven by possible military and other applications of the mechanisms for accelerated decay. Pursuit [8] of the physics of these mechanisms and the related technology is understandable because, in principle, these mechanisms might relate to possible development of new types of nuclear weapons.

In the Collins *et al.* experiments the target containing the $^{178}\mathrm{Hf}^{m2}$ isomer nuclei was irradiated with incoherent x rays. A low-intensity ("dental") x-ray apparatus with a wide spectrum of bremsstrahlung photons was used in experiments described in [1,2]. The upper frequency limit of the bremsstrahlung spectrum in [1] was 70 keV in one measurement set and 90 keV in the other. A certain intensity increase indicated triggering in the first series of measurements but was not sufficient to justify tabulation [1]. Approximately a 6% intensity increase of the 495-keV γ line [11^-(1859 keV) $\rightarrow 9^-$(1364 keV) transition] and 2% intensity increase of the 426-keV γ line [8^+(1058 keV) $\rightarrow 6^+$(632 keV) transition] was observed in the decay spectrum of the $^{178}\mathrm{Hf}^{m2}$ isomer (see Fig. 1) in the second series of measurements. The bremsstrahlung photon spectrum in Ref. [2] was cut off at 63 keV. A 1.6% increase of the 213-keV line intensity [4^+(306 keV) $\rightarrow$ 2^+(93 keV) transition] and a certain intensity increase of the 6^+(632 keV) $\rightarrow 4^+$(306 keV) transition was observed.

Results published in Ref. [6] were obtained with the synchrotron beam of the SPring-8 accelerator (Japan). Photon energies were in the range of 9–13 keV. After reaching the photoionization threshold of the L_{III} shell of Hf atoms, a 1% increase of the overall intensity of the following γ lines were observed: 213 keV [4^+(306 keV) $\rightarrow 2^+$(93 keV) transition] and 217 keV [9^-(1364 keV) $\rightarrow 8^-$(1147 keV) transition]. On reaching the L_I shell ionization threshold, the intensity increase of the 217 keV γ line was 3%.

Such intriguing results could not remain unnoticed. Comments [11–13] on article [1] were published in 2000. Theoretical estimations revealed some discrepancies between known laws of nuclear physics, including sum rules [12,13] and measured integrated cross section. Then a group of physicists from three leading U.S. research centers (Los Alamos National Laboratory, Lawrence Livermore National Laboratory, and Argonne National Laboratory) tried to observe the effect of $^{178}\mathrm{Hf}^{m2}$ induced decay in specially designed experiments [14,15]. Detailed measurements performed using the Advanced Photon Source at Argonne National Laboratory showed no intensity increase of γ transitions in ^{178}Hf nuclei irradiated with photons in the energy range of 20–60 keV [14] and 9–20 keV [15]. Note that the beam intensity in Ref. [14] was greater than that described in Collins *et al.* [1,2] by several orders of magnitude. This should have led to a significant amplification of the effect observed in Refs. [1,2]. However, the $^{178}\mathrm{Hf}^{m2}$ isomer decay rate observed in Ref. [14] remained the same within the 2% measurement error, independent of the target irradiation. As for the experiments with photons in the 9- to 13-keV energy range, the upper limit of the total cross section of the $^{178}\mathrm{Hf}^{m2}$ induced decay determined in Ref. [15] was smaller than that in experiment in Ref. [6] by three orders of magnitude. Note that the results obtained at the Argonne National Laboratory agree with those obtained at the National Synchrotron Light Source at Brookhaven National Laboratory [16]. In the latter case within the experimental accuracy no induced decay of Hf irradiated with photons in the energy range from the Hf L_I shell ionization threshold up to 12–13 keV was observed, either. All published results on triggering of $^{178}\mathrm{Hf}^{m2}$ are collected in Ref. [17].

II. MODELS OF THE INDUCED DECAY PROCESS AND THE ANALYSIS OF THE EXPERIMENTAL RESULTS

We shall try to give a simple theoretical analysis of the situation.

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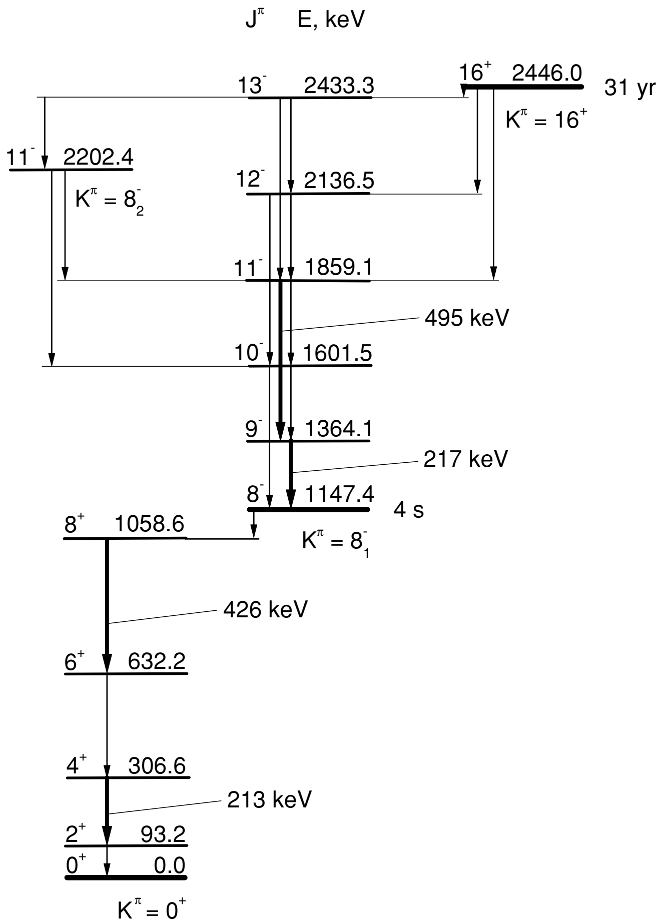


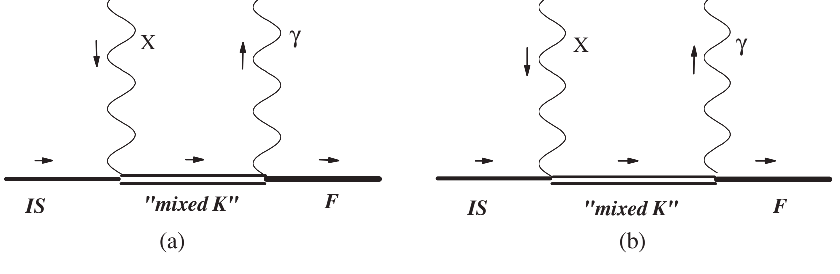
FIG. 1. ^{178}Hf^{m2} decay scheme according to Refs. [9,10]. The highlighted transitions correspond to the γ lines with intensity increase above the level of experimental error, as observed in Refs. [1,2,6].

The experimental conditions in Refs. [1,2,6] allow one to conclude that the process under consideration is purely electromagnetic in nature. No pronounced strong-field effects play any role, because the pump radiation is noncoherent and has a too low intensity. This leaves only two mechanisms for the induced decay. The first one is the direct interaction of the incident x rays with the nucleus leading to isomer decay via an intermediate state. The second is the interaction of the incident x rays with the atomic shell which, in turn, transfers the excitation to the nucleus. Diagrams describing both processes within quantum electrodynamics (QED) perturbation theory are shown in Figs. 2 and 3 (only the direct diagrams are considered, because we intend to evaluate the effect with an accuracy of an order of magnitude).

Diagrams shown in Fig. 2 describe the experiments in Refs. [1,2]. The two channels of the intermediate-state decay are shown: γ emission [Fig. 2(a)] and internal electron conversion [Fig. 2(b)]. Diagrams in Fig. 3 correspond to the experiment in Ref. [6]. The increase of the nuclear transition intensity in this experiment was observed when the L shell ionization of Hf atoms occurred. Diagrams in Fig. 3 show the nuclear excitation process due to the vacancy transfer from the inner atomic shell to the outer ones followed by the decay of the intermediate nuclear level via the same channels as in Fig. 2.

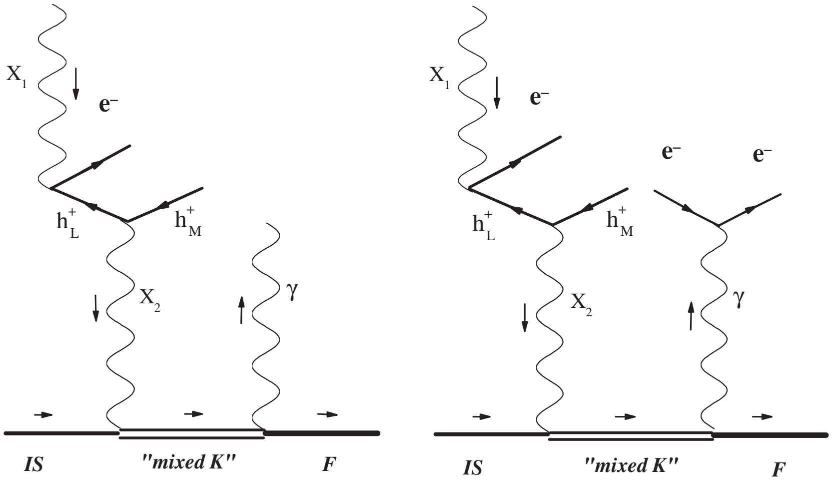
An essentially different scenario is also possible. In case of "normal" spontaneous decay the nuclear transitions from the isomeric state occur directly to the lower levels. Conditions that favor this type of nuclear transitions could be devised. This approach is considered in Sec. III of this article.

The long-lived state 16^+(2.446 MeV, 31 yr) is an interesting example of a four-quasiparticle isomer. The channels of spontaneous decay of the state 16^+(2.446 MeV, 31 yr) with $K^\pi = 16^+$ are, primarily, the $E3$ transition to the 13^-(2.433 MeV) level of the rotational band $K^\pi = 8^-$, which has the branching ratio $\beta_{E3} = 0.9982$, and the $M4$ transition to the 12^-(2.136 MeV) level of the same band with $\beta_{M4} = 0.0018$ [9]. The experimental values (in Weisskopf units) for the above-mentioned nuclear transition probabilities are as follows: $B_{\mathrm{W.u.}}(E3) = 9 \times 10^{-10}$, $B_{\mathrm{W.u.}}(M4) = 4.9 \times 10^{-8}$ [9]. The intensities of both transitions are reduced according to the known rules for K forbidden transitions [18], that result approximately in a factor of 10^2 per every unit of K forbiddenness $n = |K_i - K_f| - L$, where L is the multipolarity. [Note that the above mentioned probability of the nuclear transition $B(E/ML; i \to f)$ equals the modulus squared of the nuclear matrix element for the electromagnetic transition summed over the magnetic quantum numbers of the multipole and of the final state of the nucleus. $B_{\mathrm{W.u.}}$ is the ratio of the measured or calculated values for the $B(E/ML)$ to the reduced probability



(a) (b)

FIG. 2. QED diagrams for the decay induced by the photon-nucleus interaction.



(a) (b)

FIG. 3. Diagrams of induced decay following the inner atomic shell ionization.

in Weisskopf model $B(E/ML; W)$ [19]

$$B(EL; W) = \frac{e^2}{4\pi}\left(\frac{3}{3+L}\right)^2 R_0^{2L},$$
$$B(ML; W) = \frac{10}{(M_N R_0)^2} B(EL; W),$$

where $R_0 = 1.2A^{1/3}$ fm is the radius of the nucleus with atomic number A, e is the proton charge, and M_N is the nucleon mass.]

The induced decay cross sections for the isomer 16^+(2.446 MeV, 31 yr) measured in [1,2,6] were anomalously large. The authors of those articles explain the experimental results by the existence of a “mixed *K*” level in the excitation spectrum of ^{178}Hf nucleus that acts as an intermediate level in the induced decay process. According to the results of the experiment in Ref. [1], this level should lie about 40 ± 20 keV higher than the isomer level. And, according to the data in Ref. [6], the energy of this state is approximately 10 keV higher than the isomer energy. In all three experiments the “mixed *K*” level ensured an effective overcoming of *K* forbiddenness.

Thus, we can see that there is experimental evidence both “pro” (Refs. [1,2,6]) and “contra” (Refs. [14–16]) the induced decay effect. In this article we explore the extent to which the effect might be expected to exist from a theoretical viewpoint.

A. Decay caused by photon-nucleus interaction

Consider the induced decay caused by direct interaction of x-ray photons with the nucleus. The cross section for this process described by the diagrams in Fig. 2 can be easily calculated within QED. The wide bremsstrahlung spectrum contains photons that cause resonance excitation of certain intermediate nuclear levels. Close to resonance where the single level approximation is valid, the cross section has the Breit-Wigner form:

$$\sigma_{\text{ind}}(\omega_X) \simeq \frac{\lambda_X^2}{2\pi}\frac{\Gamma^{\text{rad}}(\omega_X; \text{IS} \to \text{“mixed } K\text{”})/2\,\Gamma^{\text{rad+conv}}(\omega_\gamma; \text{“mixed } K\text{”} \to F)/2}{(\omega_X - (E_{\text{“mixed } K\text{”}} - E_{\text{IS}}))^2 + (\Gamma^{\text{tot}}_{\text{“mixed } K\text{”}}/2)^2}. \tag{1}$$

In Eq. (1) ω_X is the energy and λ_X $(= 2\pi/\omega_X)$ is the wavelength of the x-ray quanta (the following system of units is used throughout the article: $\hbar = c = 1$). E_{IS} and $E_{\text{“mixed } K\text{”}}$ are the energies of the initial state (the isomer one in this case) and the intermediate states of the nucleus. $\Gamma^{\text{rad+conv}}(\omega_\gamma; \text{“mixed } K\text{”} \to F)$ is the sum of the radiation width Γ^{rad} and the conversion width Γ^{conv} as functions of the energy ω_γ of the nuclear transition from the intermediate “mixed *K*” level to the level *F*. $\Gamma^{\text{tot}}_{\text{“mixed } K\text{”}}$ is the total width of the intermediate state.

The radiation transition probability in Eq. (1) for the electric (*E*) or magnetic (*M*) type of the transition multipolarity *L* is determined by the following formula [19]:

$$\Gamma^{\text{rad}}(E/ML; \omega; i \to f) = 8\pi\frac{\omega^{2L+1}}{[(2L+1)!!]^2}\frac{L+1}{L}B(E/ML; i \to f). \tag{2}$$

We use the well-known representation of the δ function as the limit of the deltalike Cauchy sequence [20]. After substitution of the energy denominator in Eq. (1) near the resonance with $2\pi/\Gamma^{\text{tot}}_{\text{“mixed } K\text{”}}\delta\,[\omega_X - (E_{\text{“mixed } K\text{”}} - E_{\text{IS}})]$, the integral cross section [Eq. (1)] has the following form:

$$\int \sigma_{\text{ind}}(\omega_X)d\omega_X \simeq \frac{\lambda_{X_r}^2}{4}\Gamma^{\text{rad}}(\omega_{X_r}; \text{IS} \to \text{“mixed } K\text{”}) \times \frac{\Gamma^{\text{rad+conv}}(\omega_\gamma; \text{“mixed } K\text{”} \to F)}{\Gamma^{\text{tot}}_{\text{“mixed } K\text{”}}}. \tag{3}$$

Here the energy of the resonance photons is already determined by the condition $\omega_{X_r} = E_{\text{“mixed } K\text{”}} - E_{IS}$.

Preliminary estimates have shown that the cross sections measured in the experiments [1,2,6] significantly exceed those that can be obtained theoretically without involving any extraordinary ideas. Therefore, following the arguments of Collins *et al.* [1–7], let us introduce a certain additional level into the ^{178}Hf excitation spectrum and suppose that this level has a number of unusual properties. (a) Let us “forget” about the K forbiddenness and assume that for unknown reasons the intermediate level is connected both with the isomer state and all other levels of the rotational bands via K-allowed transitions. (b) Furthermore, let us suppose that the intermediate nuclear level has spin 15^- so that an $E1$ transition between this level and the 16^+(2.446 MeV) isomer is possible. (The level with spin 17^- is less suitable for explanation of the experimental results. This is clarified below.) (c) We also suppose that the nuclear matrix element of the above-mentioned $E1$ transition has the maximum value possible for the ^{178}Hf nucleus; that is, it equals the matrix element of the collective transition to the giant dipole resonance (GDR) state. And, finally, (d) a similar assumption is made for all the transitions from the intermediate state to the lower lying levels. That reduces the probability of the nucleus returning to the isomer level. Thus these assumptions provide conditions for the maximum population of the states that correspond to the transitions with increased intensity observed in Refs. [1,2,6].

The reduced probability $B(E1_{\text{GDR}})$ can be calculated using the assumption that the transition from the isomer level to the corresponding giant resonance state fully exhausts the classical dipole sum rule $S(E1)$ [18]. The equation

$$E_{\text{GDR}}B(E1_{\text{GDR}}) = \frac{9e^2}{4\pi}\frac{1}{2M_N}\frac{NZ}{A},$$

where $E_{\text{GDR}} = \varepsilon_{E1}A^{-1/3}$ is the GDR energy, the parameter $\varepsilon_{E1} \simeq 78$ MeV, and Z and N are the numbers of protons and neutrons in the nucleus, respectively, yields the following value for the probability: $B_{\text{W.u.}}(E1_{\text{GDR}}) \simeq 22.5$. The greatest value of the radiation width of the transition between the isomer level and the intermediate level lying at the distance of 40 keV would be $\Gamma^{\text{rad}}(E1_{\text{GDR}}; \text{IS} \to \text{“mixed } K\text{”}; 40 \text{ keV}) \simeq 3 \times 10^{-3}$ eV. Similar estimates for the nuclear reduced probabilities can be easily done based on the rules of the sum from Ref. [18], also for transitions with multipolarity $L \geqslant 2$.

To detect the induced decay, the probability of the nucleus returning to the isomer state from the intermediate state should be less than 1. According to the scheme in Fig. 1, the induced process probability is greater in case of a state with spin 15^- compared to the case of spin 17^-. Among the possible final states F, the states 13^-(2.433 MeV), 12^-(2.136 MeV), and 11^-(1.859 MeV) of the $K^\pi = 8_1^-$ band are of most interest. Population of these states causes the probability to increase for the transitions discussed in Refs. [1,2,6]. Population of the levels in other bands during the decay of the intermediate state has practically no influence on the intensity of transitions in the lower part of the $K^\pi = 8_1^-$ band. This is due to the fact that further in the interband transitions either K forbiddenness plays a role or the transition multipolarity is high (for the levels belonging to the bands with neighbor values of K). The population of the $K^\pi = 8_2^-$ band plays no role, either: the transition from the 11^-(2.202 MeV) state to the 11^-(1.859 MeV) state shown in Fig. 1 is weaker than the intraband $E2$ transition 13^-(2.433 MeV)→ 11^-(1.859 MeV) by three orders of magnitude [10].

The calculations show that the population of the nearest state 13^-(2.433 MeV) is the optimal for the isomer 16^+(2.446 MeV) induced decay. That ensures the $E2$ transition (this is the lowest multipolarity that is possible) from the intermediate level 15^- with the radiation width $\Gamma^{\text{rad}}(E2_{\text{GQR}}; \omega_\gamma = 53 \text{ keV}) \simeq 10^{-8}$ eV and conversion coefficient $\alpha \simeq 60$. (GQR in the expression for the Γ^{rad} means that the matrix element of the transition to the state of giant quadrupole resonance was used.) The intermediate level decay to the levels 12^-(2.136 MeV) and 11^-(1.859 MeV) via the radiation and conversion channels gives less than 1% of the probability compared to that of the transition to the 13^-(2.433 MeV) level. The reason for this is the lower intensity of the γ and conversion transitions with higher multipolarity L. The probability of γ radiation is proportional to $\omega(R/\lambda)^{2L}$ and the factor $R/\lambda \ll 1$ in the considered energy range. The relative increase of the conversion coefficients with the growth of L cannot compensate for the decrease of the radiation width.

All the above considerations lead to the following upper limit for the integral cross section within assumptions (a)–(d):

$$\int \sigma_{\text{ind}}(\omega_X)d\omega_X \leqslant 10^{-27} \text{ cm}^2 \text{ keV}.$$

That is still less than the value ($\sim 10^{-21}$ cm^2 keV) measured in Ref. [1] by six orders of magnitude. The only way to compensate for this discrepancy is to assume the existence of a “continuum,” that is, approximately 10^6 of nonoverlapping intermediate levels lying 40 ± 20 keV above the isomer state and possessing the unlikely assumptions (a)–(d).

It is easy to see that there is a certain “reserve” in Eq. (3) for the increase of the cross-section theoretical value. We add yet another assumption to the four discussed above. (e) We assume that there is a level in the spectrum of ^{178}Hf below the intermediate 15^- “mixed K” state, such that the partial width of transition to this level almost exhausts the total width of the intermediate state $\Gamma^{\text{tot}}_{\text{“mixed } K\text{”}}$. (This hypothesis is implied in [6], as discussed in Sec. II B of the present article.) Now assuming that in Eq. (3):

$$\frac{\Gamma^{\text{rad+conv}}(\omega_\gamma; \text{“mixed } K\text{”} \to F)}{\Gamma^{\text{tot}}_{\text{“mixed } K\text{”}}} \sim 1, \tag{4}$$

we obtain the following estimate for the integral cross section:

$$\int \sigma_{\text{ind}}(\omega_X) d\omega_X \leqslant 10^{-23}\ \text{cm}^2\ \text{keV}.$$

We can see that even in this case the estimated value is two orders of magnitude less than the measured one.

In Ref. [2] the following estimate is given for the integral cross section of induced decay via the intermediate "mixed *K*" level lying $\leqslant$20 keV above the isomer level: 2.2×10^{-22} cm^2 keV. A theoretical estimate of the cross section for this level would be also different. The radiation width of the $E1$ transition (2) is proportional to the third power of energy. Therefore substitution of 40 keV with 20 keV in this formula would give the following inequalities:

$$\int \sigma_{\text{ind}}(\omega_X) d\omega_X \leqslant 10^{-28}\ \text{cm}^2\ \text{keV}.$$

within the frame of assumptions (a)–(d) and

$$\int \sigma_{\text{ind}}(\omega_X) d\omega_X \leqslant 10^{-24}\ \text{cm}^2\ \text{keV}.$$

with the additional assumption (e). This is still less than the measured values by six and two orders of magnitude, respectively.

B. Decay due to the photon interaction with the atomic shell

In this section we analyze Ref. [6]. In this experiment, an enhancement of the isomer 16^+(2.446 MeV, 31 yr) spontaneous decay by 1–3% accompanying the atomic *L* shell ionization of Hf atoms with synchrotron radiation with photon flux density $\varphi \simeq 10^{11}$ cm^{-2} s^{-1} was observed. The measured enhancement $f \simeq 0.01$–0.03 allows one to calculate the cross section of the induced decay as follows: $\sigma_{\text{ind}} = f \ln 2/(T_{1/2}^{\text{IS}}\varphi) \simeq 2 \times 10^{-22}$ cm^2. The authors assumed that the observed effect was due to the excitation of the ^{178}Hf nucleus from the isomer state to the intermediate "mixed *K*" level due to the transition of one of the atomic electrons to the hole in *L* shell. Using the known photoionization cross section $\sigma_{\text{ion}}^{(L)} \simeq 7.5 \times 10^{-20}$ cm^2 [21], the authors estimated the probability *P* of this process as follows:

$$P = \sigma_{\text{ind}} \big/ \sigma_{\text{ion}}^{(L)} \simeq 2 \times 10^{-3}.$$

Note that the formula contains no branching ratio. And this is equivalent to this ratio being 1. The latter, as we have shown, is possible only in case of a level being present in the spectrum below the intermediate "mixed *K*" state with properties given by assumption (e).

Further, we analyze the value of $\sigma_{\text{ind}} \simeq 2 \times 10^{-22}$ cm^2 measured in Ref. [6], first using the assumptions (a)–(d) and then adding the assumption (e).

Assume the resonant excitation of the intermediate "mixed *K*" level due to an $E1$ transition of the electron from the M_{II} to the L_I atomic shell of the hafnium atom. (The choice of the subshells ensures the largest atomic matrix element [22].) Note that in this case the "mixed *K*" level lies approximately 8.9 keV above the isomer state 16^+(2.446 MeV, 31 yr). For the process under consideration the assumption of the resonant character of the excitation is by far not so trivial as in case of photoexcitation with bremsstrahlung. Now the transition between the intermediate and the isomer levels should have the energy ω_N equal to the atomic transition energy ω_A within the vacancy width $\Gamma_{L_I} + \Gamma_{M_{II}} \simeq 10$ eV [23]. Only a few cases of such unique matching are known. Therefore we call this very strong assumption (f).

The theory of nuclear excitation by electron transition (NEET), developed in Refs. [24–26], with the correction suggested in Ref. [27], gives results that are in agreement with modern experimental data [28–31]. Therefore, this theory can be used in the analysis of the results in Ref. [6].

The relative probability of nuclear excitation with a virtual photon emitted by an atomic electron is determined by the following formula [24–26]:

$$P_{\text{NEET}} = \left(1 + \frac{\Gamma_{M_{II}}}{\Gamma_{L_I}}\right) \frac{E_{\text{int}}^2}{(\omega_N - \omega_A)^2 + \left(\Gamma_{L_I} + \Gamma_{M_{II}}\right)^2/4}. \tag{5}$$

The key parameter in Eq. (5) is the interaction energy E_{int} of the electron $j^{\mu}_{L_I M_{II}}(\mathbf{r}) = -e\bar{\psi}_{L_I}(\mathbf{r})\gamma^{\mu}\psi_{M_{II}}(\mathbf{r})$ and nuclear $J^{\nu}_{\text{“mixed }K\text{” IS}}(\mathbf{R}) = e\Psi^{+}_{\text{“mixed }K\text{”}}(\mathbf{R})\hat{J}^{\nu}\Psi_{\text{IS}}(\mathbf{R})$ electromagnetic currents of the transition. E_{int}^2 is the modulus squared of the interaction Hamiltonian H_{int} averaged over the initial states and summed over the final states as follows:

$$H_{\text{int}} = \int d^3r d^3R\, j^{\mu}_{L_I M_{II}}(\mathbf{r}) D_{\mu\nu}(\omega_N; \mathbf{r} - \mathbf{R}) J^{\nu}_{\text{“mixed }K\text{” IS}}(\mathbf{R}), \tag{6}$$

where $D_{\mu\nu}(\omega_N; \mathbf{r} - \mathbf{R})$ is the photon propagator. The numeric computation for the transition complying with the assumptions (a) and (b) of the model introduced in Sec. II A yields $E_{\text{int}} \simeq 1.3 \times 10^{-1}$ eV. This is the largest possible energy that can cause the nuclear transition IS → "mixed *K*" accompanying the electron transition to the *L* shell.

The probability P_{NEET} under the assumptions (a)–(d) is $\simeq 1.1 \times 10^{-3}$. The calculation of the branching ratio $\beta_{\text{“mixed }K\text{”}}$ for the same model was given in Ref. [22]. The result is $\beta_{\text{“mixed }K\text{”}} < 10^{-3}$. Therefore the following estimate for the induced decay cross section is valid:

$$\sigma_{\text{ind}} < 10^{-25}\ \text{cm}^2.$$

So the cross section is at least three orders of magnitude less than that measured in Ref. [6].

Let us add assumption (e). Even then the upper limit of the calculated cross section of the isomer induced decay is 4–5 times less than the experimental value $(1.77$–$1.95) \times 10^{-22}$ cm^2. In addition to that, the probability of the simultaneous realization of assumptions (a)–(f) appears to be extremely low, because even the separate assumptions look completely unreal.

III. DECAY IN THE INVERSE NEET PROCESS

Below we verify the last hypothesis, namely that the lifetime decrease of the isomer 16^+(2.446 MeV, 31 yr) is due to the probability increase of the $E3$ transition with energy 12.7 keV to the 13^- level (2.433 MeV). This nuclear transition is the principal channel for the isomer decay in normal conditions.

Such an acceleration could be due to the interaction with the atomic shell via an inverse process of nuclear excitation by electronic transitions (NEET) [32]. This scenario is possible, for example, for the isomer level decay in ^{197}Au, ^{193}Ir [32], and other nuclei, if the outer electron shell that participates in the NEET process is ionized. It is also shown in Ref. [32] that in the resonance case this process ensures the maximum possible enhancement of decay compared to all the other processes where the nucleus interacts with the electronic shell.

To obtain the upper limit of the process cross section we make several assumptions that ensure the optimal conditions for decay via this channel. We assume first that after L shell ionization of the Hf atom, ionization of the outer atomic shells occurs with unit probability in the electron transition $M \rightarrow L$ (Auger process), so that the distance between the states L_{III} and M_{IV} becomes exactly equal to the energy of the $E3$ nuclear transition $16^+(2.446\ \text{MeV},\ 31\ \text{yr}) \rightarrow 13^-(2.433\ \text{MeV})$. Assume also that the L_{III} shell is populated and the M_{IV} shell is depopulated. Thus the conditions for the resonance inverse NEET process (INEET) are ensured. The probability of this process is calculated using the formula $W_{\text{INEET}} = \Gamma_{M_{IV}} P_{\text{INEET}}$ [32], where P_{INEET} is the relative probability of atom excitation from the L_{III} state to the M_{IV} state because of the considered nuclear transition. This probability is estimated from Eq. (5) with corresponding substitutions of the atomic and nuclear initial and final states. The interaction energy $E^2_{\text{int}}(E3; M_{IV} \rightarrow L_{III}; \text{IS} \rightarrow 13^-)$ that initiates the INEET process is $1.3 \times 10^{-23}\ \text{eV}^2$. Such a small value is because of the K forbiddenness of the $E3$ nuclear transition. The relative probability of INEET is also very small: $P_{\text{INEET}} \simeq 6 \times 10^{-24}$.

The number of isomeric nuclei that could have decayed via the INEET channel per unit time Q_{INEET} in the experiment in Ref. [6] can be estimated with the following relation:

$$Q_{\text{INEET}} \simeq N_{\text{IS}} P_{\text{INEET}} \sigma^{(L)}_{\text{ion}} \varphi_X,$$

where N_{IS} is the number of isomeric hafnium nuclei in the target. Comparing this rate to the natural rate of isomer decay $Q = \lambda_{\text{IS}} N_{\text{IS}}$ (here $\lambda_{\text{IS}} = \ln 2/T^{\text{IS}}_{1/2}$), we obtain the following:

$$\frac{Q_{\text{INEET}}}{Q} \simeq \frac{P_{\text{INEET}} \sigma^{(L)}_{\text{ion}} \varphi_X}{\lambda_{\text{IS}}} \simeq 2 \times 10^{-23}.$$

This result excludes the possibility of explaining the experimental data Ref. [6] with the enhancement of the $E3$ nuclear transition $16^+(2.446\ \text{MeV},\ 31\ \text{yr}) \rightarrow 13^-(2.433\ \text{MeV})$ because of the interaction with the atomic shell. No assumptions about the properties of the nuclear transition [similar to assumptions (a)–(d)] can be used here. The intensity of the transition is measured and leaves no room for speculation.

IV. DECAY IN AN INTENSIVE EXTERNAL FIELD

The existence of a "mixed K" state should lead to a number of observable effects. Decay acceleration in an intensive external field is among them. Laser radiation could be used as the source of such a field.

Consider the inelastic scattering of optical photons with $^{178}\text{Hf}^{m2}$ nuclei. The diagrams of this process are shown in Fig. 2. Decay induced by interaction with laser photons differs somewhat from the process discussed in Sec. II A. In the present case we are dealing with monochromatic radiation and a nonresonant process.

The formula for the probability of isomer level induced decay in the field of laser radiation with intensity I_L in single-level approximation is similar to the one in Eq. (1):

$$W_{\text{ind}} \simeq \frac{I_L}{\omega_L} \frac{\lambda_L^2}{8\pi} \frac{\Gamma^{\text{rad}}(\omega_L; \text{IS} \rightarrow \text{“mixed } K\text{”})\, \Gamma^{\text{rad+conv}}(\omega_\gamma; \text{“mixed } K\text{”} \rightarrow F)}{(E_{\text{“mixed } K\text{”}} - E_{\text{IS}})^2}, \tag{7}$$

where ω_L is the photon energy. This formula also accounts for the fact that $\omega_L \ll E_{\text{“mixed } K\text{”}} - E_{\text{IS}}$ and $\omega_\gamma \simeq E_{\text{IS}} - E_F$. The validity of a single-level approximation is based on the properties of the intermediate "mixed K" level: the transition matrix element to the "mixed K" state is greater than those of transitions to other nuclear states by many orders of magnitude.

The physics underlying the induced decay described by Eq. (7) is fairly simple. We substitute the expression for an electric dipole transition in the Weisskopf model from Eq. (2) $\Gamma^{\text{rad}}(E1; \omega_L) = e^2 \omega_L^3 R_0^2/4$ into this formula. Taking into account the following relations: $I_L = \mathcal{E}^2/4\pi$ (where $\mathcal{E}$ is the electric field strength) and $d = eR_0$ (where d is the dipole moment), we obtain the following expression:

$$W_{\text{ind}} \sim a^2_{\text{“mixed } K\text{”}} \Gamma^{\text{rad+conv}}(\omega_\gamma; \text{“mixed } K\text{”} \rightarrow F), \tag{8}$$

where

$$a_{\text{“mixed } K\text{”}} = \frac{\mathcal{E} d}{E_{\text{“mixed } K\text{”}} - E_{\text{IS}}}. \tag{9}$$

[In case of a very strong field the conversion channel of decay could be closed due to ionization of the atomic shells, and only the radiation part of transition width would remain in Eq. (8).]

It is well known that the wave function (WF) Ψ_{IS} of the isomer state in a strong external field is transformed into a new WF Ψ [33]. Wave function Ψ is no longer an eigenfunction of the initial nuclear Hamiltonian. An admixture of other states is added to the function Ψ_{IS} because of interaction with the external field. The state Ψ_n connected by the electric dipole transition with the function Ψ_{IS} gives the main contribution to this admixture in an electric field

$$\Psi \simeq \Psi_{\text{IS}} + a_n \Psi_n + \cdots. \tag{10}$$

Note that factor a_n in Eq. (10) is the same as the similar factor in Eq. (9). Thus an additional channel for the isomeric decay in an external field via the admixture of an intermediate level appears. If the multipolarity of the $n \rightarrow F$ transition is less than that of the isomeric transition, then the small value of factor a_n is compensated for by the greater probability of the admixture decay [34].

Below we estimate the laser radiation intensity necessary to increase the probability of isomer $16^+(2.446\ \text{MeV},\ 31\ \text{yr})$ decay by a factor of 2, if the measurements in Ref. [1,2] are accurate or the model formulated in assumptions (a)–(d) of Sec. II A are valid. [Here the following should be noted. As was shown above, assumptions (a)–(d), introduced in Sec. II A concerning the properties of the intermediate "mixed K" level, are insufficient to reproduce the experimental results [1,2].

It is necessary to have not just a single "mixed K" level but approximately 10^6 of such intermediate levels. During the excitation of ^{178}Hf^{m2} with a wide bremsstrahlung spectrum no interference of the states occurs, because different resonance photons correspond to the different nonoverlapping states, so that their excitations occurs independently. In the case of induced decay in a strong external field, however, the process is not the same. The wave function described in Eq. (10) contains the admixtures of all the 10^6 states. The calculation of cross section or transition probabilities includes interference components that cannot be deduced from the data of Refs. [1,2]. Therefore the estimate of W_{ind} in Eq. (8) based on the data in Refs. [1,2] should be performed with care.]

W_{ind} is independent of the wavelength of optical photons in Eqs. (7) and (8). Everything depends on the laser radiation intensity, the value of the matrix element for the $E1$ transition from the isomer state to the intermediate level, the width $\Gamma^{\mathrm{rad+conv}}(\omega_\gamma = E_{\mathrm{IS}} - E_F;$ "mixed K" $\to F)$, and the energy denominator $\Delta = E_{\text{“mixed }K\text{”}} - E_{\mathrm{IS}}$. Within the considered model [see Sec. II A assumptions (a)–(d)], $\Gamma^{\mathrm{rad}}[E2_{\mathrm{GQR}}; \omega_\gamma = 12.7$ keV; "mixed K" $\to 13^-(2.443\ \mathrm{MeV})] \simeq 5 \times 10^{-12}$ keV. The conversion coefficient for this transition, according to our calculations, is $\alpha \simeq 0.7 \times 10^5$. Assume also that $\Delta \simeq 10$–20 keV.

Then W_{ind} in Eq. (7) reaches the value equal to that of the spontaneous isomer decay $W_{\mathrm{IS}} = 7.1 \times 10^{-10}\ \mathrm{s}^{-1}$ at a laser intensity of $I \simeq 10^{12}\ \mathrm{W\ cm}^{-2}$. (The electric field strength corresponding to this intensity is $\mathcal{E} \simeq 10^8\ \mathrm{V\ cm}^{-1}$. No full ionization of the L, M shells of a Hf atom occurs at such field strengths, so the conversion channel is still open.) This result is valid for the case of a single "mixed K" level. In the case of 10^6 intermediate "mixed K" states in the spectrum the absence of interference decreases the laser intensity necessary for this process to a value of approximately $10^6\ \mathrm{W\ cm}^{-2}$. Destructive interference can turn the relative probability of the induced process to zero, and constructive interference can lead to isomeric decay enhancement by a factor of 2 already at the intensity of $I \simeq 1\ \mathrm{W\ cm}^{-2}$. This allows a qualitative verification of the experimental results [1,2] using laser radiation.

V. FURTHER COMMENTS

There are several remarks.

1. The measurement procedures in Refs. [14,15] and [1,2,6] are different. In Refs. [14,15] the presence or absence of induced decay of the isomer $16^+(2.446$ MeV, 31 yr) is determined by the change of decay intensity of another isomer level, namely $8^-(1.147$ MeV, 4 s). [It should be noted that the integrated cross section limits shown in Fig. 4 from Ref. [15] are for photon-induced deexcitation of the $16^+(2.446$ MeV, 31 yr) isomer either through the 8_1^- isomer or for decay that somehow bypasses the 8_1^- isomer.] Measurements of the 8_1^- isomer decay are quite natural, because the Collins group has determined the intensity increase for the transitions with energies 495 and 217 keV (see Fig. 1). Nevertheless, it is often argued that the intermediate "mixed K" level can decay directly to the states of the $K^\pi = 0^+$ band and not via the isomer $8^-(1.147$ MeV, 4 s).

We verify this possibility and estimate the probabilities of population of levels in this band via transitions from the 15^- "mixed K" state (below referred to as 15^-). We also compare these probabilities to the width $\Gamma^{\mathrm{rad+conv}}[E2$, 15^- "mixed K" $\to 13^-(2.433\ \mathrm{MeV})]$, hereafter designated as $\Gamma^{\mathrm{tot}}(E2; 15^- \to 13^-)$. There are only two levels in the $K^\pi = 0^+$ band below the "mixed K" level in addition to those shown in Fig. 1, namely $10^+(1.571$ MeV) and $12^+(2.151$ MeV) [9]. A simple calculation shows that the following relations are valid under assumptions (a)–(d): $\Gamma^{\mathrm{rad+conv}}[E3, 15^- \to 12^+(2.151\ \mathrm{MeV})]/\Gamma^{\mathrm{tot}}(E2; 15^- \to 13^-) \leqslant 10^{-3}$, $\Gamma^{\mathrm{rad+conv}}[E5, 15^- \to 10^+(1.571\ \mathrm{MeV})]/\Gamma^{\mathrm{tot}}(E2; 15^- \to 13^-) \leqslant 10^{-9}$, $\Gamma^{\mathrm{rad+conv}}[E7, 15^- \to 8^+(1.058\ \mathrm{MeV})]/\Gamma^{\mathrm{tot}}(E2; 15^- \to 13^-) \leqslant 10^{-16}, \ldots$ $\Gamma^{\mathrm{rad+conv}}[E11, 15^- \to 4^+(306\ \mathrm{keV})]/\Gamma^{\mathrm{tot}}(E2; 15^- \to 13^-) \leqslant 10^{-31}$, and so on. In the Weisskopf model the population of the $K^\pi = 0^+$ band is still less than that. High multipolarity for $R/\lambda \ll 1$ suppresses the transitions.

The same is also true for the levels in other rotational bands, because we assume the absence of K forbiddenness for all transitions from the "mixed K" level and take into account only the spins of the nuclear states. According to Ref. [9], the levels belonging to the rotational bands of the ^{178}Hf nucleus, lying below the isomer $16^+(2.446$ MeV, 31 yr) and not shown in the diagram in Fig. 1, have spins $\leqslant 10$. For example, the $K^\pi = 1^-$ band with head level 1^- (1.310 MeV) discussed in [7] cannot be used instead of the $K^\pi = 8_1^-$ band to explain the intensity enhancement of the 216- to 217-keV transition in the induced decay process via the intermediate "mixed K" level.

Thus the available data concerning the excitation spectrum of ^{178}Hf allows one to conclude that the isomeric level $8^-(1.147$ MeV, 4 s) is inevitably populated in the process of "mixed K" state decay.

Adding in the diagram an arrow pointing from the intermediate "mixed K" level to somewhere down to the lower lying levels can be used to "explain" the absence of certain lines in γ spectrum [7]. However, this method cannot validate the necessary cross-section value. This has been shown in Sec. II A, where an ideal partner level for the intermediate state decay was modeled using assumption (e) and Eq. (4).

2. Nuclei decay diagrams are refined as more advanced measurement techniques are developed and device sensitivity is enhanced. Sometimes new transitions (mostly low intensity) and new levels (mostly weakly populated) are added to these diagrams. Refs. [1–7] claim to validate quite a different scheme. "Mixed K" states from Refs. [1–7] should have such unusual properties for validation of the measurement cross-section value that they could change known decay schemes and γ spectra of some nuclei appreciably. Until now there are no other grounds for such a change but the results of the several experiments of Collins *et al.*

VI. CONCLUSION

To summarize, it should be stated that measurements cited in Refs. [1,2,6] are incompatible with the well-founded modern conceptions of the physics of atomic nuclei and the nature of electromagnetic processes in nuclei. Introduction of new exotic nuclear levels cannot change the situation in general.

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