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Induced decay of the nuclear isomer ^{178m2}Hf and the 'isomeric bomb'

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1. Introduction

Recently, there have been reports in the mass media about plans to build what became known as an 'isomeric bomb' based on ^{178}Hf [1]. What all the publications are speaking about is no less than the possibility of building a radically new weapon that does not fall under a single article of the existing nonproliferation treaties. The publications were based on the sensational results on induced decay of the long-lived isomer ^{178m2}Hf(16^+, 2446 keV, 31 yr) [2–10], obtained in 1999–2004 by a group of researchers headed by Carl B Collins, the Director of the Center for Quantum Electronics, University of Texas at Dallas. Despite the five-year history of the issue, so far there have been no scientific publications on this topic in Russia. The present report is an attempt to fill this gap.

A substance with stored energy and a physical process that ensures the rapid liberation of this energy are two components of any explosive device. In the case of a 'hafnium' bomb, the energy is stored in a metastable state and amounts to 2.446 MeV per nucleus, or 1.3 GJ per gram of substance. In the opinion of Pentagon experts [11], "such extraordinary energy density has the potential to revolutionize all aspects of warfare." The only question is how to ensure the controllable decay of ^{178m2}Hf.

A simple way of accelerating the decay of the isomer was developed in the experiments of the Texas Collaboration [2, 3]. A target containing ^{178m2}Hf was subjected to the radiation

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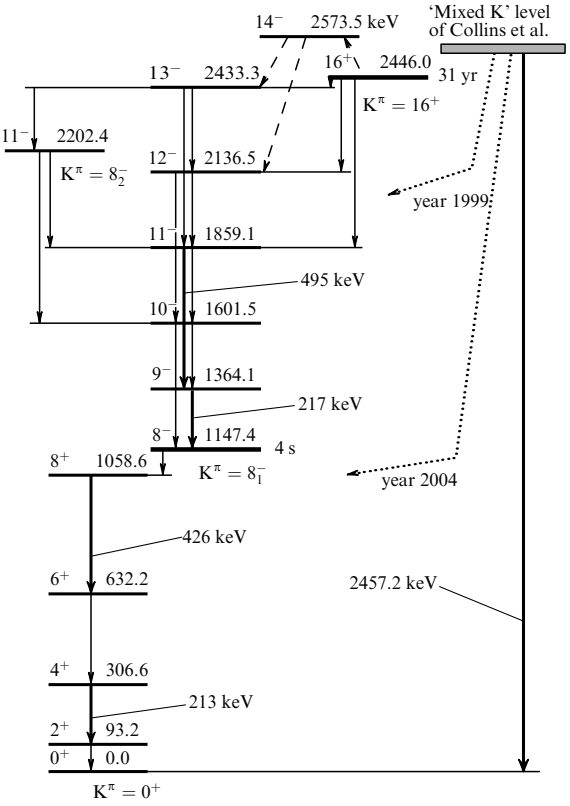

Figure 1. Decay of ^{178m2}Hf according to the data in Refs [12, 13]. Depicted are transitions in the spectrum of ^{178}Hf in which, according to Refs [2, 3, 7], an increase in the gamma-radiation intensity exceeding the measurement errors and the 2457.2 keV line discovered by Collins et al. [10] were detected. The dotted lines show the evolution of the Texas Collaboration ideas in Refs [2–10] concerning the decay of intermediate 'mixed K' states from Ref. [2] published in 1999 to Ref. [10] published in 2004. The dashed lines show the scheme of the possible experiment in the induced decay of the isomer through the 14^-(2573.5 keV) level (see Section 4 of the present paper).

emitted by a dental X-ray unit. The upper edge of the photon spectrum in the experiment described in Ref. [2] was 70 keV in one set of measurements and 90 keV in another. In the first case, no statistically significant increase in the intensity of gamma transitions was recorded, but in the second case, a 6% increase in the intensity of the 495 keV gamma line and a 2% increase in the intensity of the 426 keV gamma line were recorded in the decay spectrum of the isomer ^{178m2}Hf (Fig. 1). In the experiment described in Ref. [3], the electron bremsstrahlung spectrum was cut off at 63 keV, and a 1.6% increase in the intensity of the 213 keV gamma line was recorded.

The experiments described in Refs [7, 10] were conducted with the synchrotron radiation beam on the SPring-8 accelerator (Japan). In the first experiment (see Ref. [7]), the photon energies were varied from 9 to 13 keV. As the photoionization threshold for the L_{III}-shell of the hafnium atom was reached, a 1% increase in the total intensity of the 213 keV and 217 keV gamma lines was recorded. As the photoionization threshold for the L_I-shell of the hafnium atom was reached, the increase in the intensity of the 213 keV gamma line amounted to 3%. In the second experiment (see Ref. [10]), all photons with energies higher than 100 keV were recorded. When the energy of the synchrotron radiation reached 9567 eV, then, first, the overall count of such photons was found to increase by 3.6 to 5% and, second, a

line at 2457.2 keV new for the ^{178}Hf nucleus, appeared. In addition to this, a line at 130.2 keV was detected [9], which is uncharacteristic for the emission spectrum of ^{178}Hf.

Some of the results of the Texas Collaboration (see Refs [2, 3, 7, 10]) have already been checked. Already in the first responses [14 – 16] to Ref. [2], it was noted that the measured value of the integrated cross section does not agree with a number of well-established facts and, in particular, with the sum rule [15, 16]. Then a group of physicists from the Los Alamos National Laboratory, the Lawrence Livermore National Laboratory, and the Argonne National Laboratory tried to detect the effect of induced decay of ^{178m2}Hf in specially designed experiments [17, 18]. Detailed measurements done with the Advanced Photon Source at the Argonne National Laboratory detected no increase in the intensity of gamma transitions in the nucleus of ^{178}Hf when the target was irradiated by photons in the 20 – 60 keV range [17] and in the 9 – 20 keV range [18]. Incidentally, in the experiment described in Ref. [17], the intensity of the photon flux was several orders of magnitude higher than that of bremsstrahlung X rays in the works of the Texas Collaboration [2, 3]. Accordingly, one could expect a substantial increase in the effect against that observed by the collaboration (see Refs [2, 3]). However, the rate of decay of the isomer ^{178m2}Hf in Ref. [17] remained the same (to within a 2% measurement error) irrespective of whether the target was irradiated. In the experiments with 9 – 13 keV photons, too, the upper limit for the integrated cross section of induced decay of ^{178m2}Hf established in Ref. [18] was found to be roughly a thousand times smaller than the value obtained in the experiments of the Texas Collaboration [7]. We note that the result obtained with the synchrotron at the Argonne National Laboratory agrees with the results of measurements performed by Roberts et al. [19] with the beam of the National Synchrotron Light Source at the Brookhaven National Laboratory, who were also unable to confirm (within measurement errors) the presence of induced decay caused by photons in the energy range from the ionization potential of the L_I-shell of the hafnium atom to 12 – 13 keV. Finally, Carroll et al. [20] arrived at a negative result in their experiment with bremsstrahlung with a 100 keV ultimate energy of the bremsstrahlung photons.

Results of the experiments performed in 1998 – 2003 are presented systematically in Ref. [21].

2. Models of the induced decay process and analysis of the experimental results

We try to clarify the above situation by doing a simple theoretical analysis.

A careful study of the conditions of the experiments described in Refs [2, 3, 7, 10] reveals that we are dealing solely with electromagnetic processes and that, obviously, there are no effects related to strong electromagnetic fields, because the incident radiation is incoherent and its intensity is not high enough. This situation presents only two possibilities for induced decay. First, the X-ray radiation leaves the atomic shell intact and acts directly on the nucleus, causing the isomer to decay through an intermediate nuclear level. Second, X-ray photons interact with the atomic shell, and the excitation is passed to the nucleus from this shell. Diagrammatic representations describing both processes in the QED perturbation theory setting are given in Figs 2 and 3 (we limit ourselves to direct diagrams, because our aim here is to estimate the effect's order of magnitude).

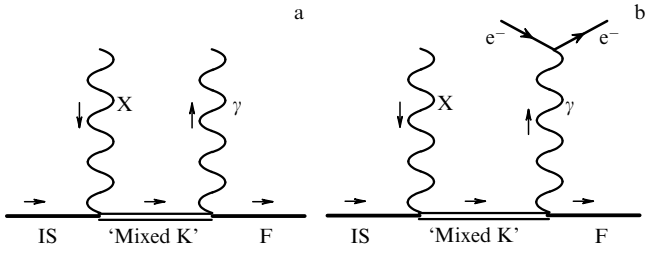

Figure 2. Diagrams of induced decay with the photon – nucleus interaction. The following channels of decay of the intermediate nuclear state are depicted: (a) gamma-ray emission and (b) internal electron conversion.

There is also a third, quite different, scenario. Under 'normal' spontaneous decay, nuclear transitions proceed from the isomer state directly to lower levels. Presumably, conditions can be created in which the probability of one of these nuclear transitions increases. This variant is discussed in Section 2.3 below.

2.1 Decay in the presence of photon – nucleus interaction

We examine the induced decay when the X-ray photons interact directly with the nucleus. The cross section of the process described by the sum of the diagrams in Fig. 2 can easily be calculated in accordance with the QED rules. The broad bremsstrahlung spectrum contains photons that resonantly excite one intermediate nuclear level or another.

Near a resonance, where the single-level approximation works, the cross section can be calculated by the Breit – Wigner formula

$$\sigma_{\rm ind}(\omega_{\rm X}) \simeq \frac{\lambda_{\rm X}^2}{2\pi}\,\frac{\Gamma^{\rm rad}(\omega_{\rm X};\,{\rm IS}\to\text{`mixed K'})/2\,\Gamma^{\rm rad+conv}(\omega_\gamma;\,\text{`mixed K'}\to{\rm F})/2}{\left[\omega_{\rm X}-(E_{\text{`mixed K'}}-E_{\rm IS})\right]^2+(\Gamma^{\rm tot}_{\text{`mixed K'}}/2)^2}\,. \tag{1}$$

Here, $\omega_{\rm X}$ is the energy, $\lambda_{\rm X}$ $(=2\pi/\omega_{\rm X})$ is the wavelength of the X-ray photons (the system of units where $\hbar=c=1$ is adopted in this report), $E_{\rm IS}$ and $E_{\text{`mixed K'}}$ are the energy of the initial state (in the case considered, the isomer state) and the energy of the intermediate state of the nucleus, $\Gamma^{\rm rad+conv}(\omega_\gamma;\,\text{`mixed K'}\to{\rm F})$ is the sum of the radiation width ($\Gamma^{\rm rad}$) and the conversion width ($\Gamma^{\rm conv}$) as functions of the energy ω_γ of the nuclear transition from the intermediate level to the level F, and $\Gamma^{\rm tot}_{\text{`mixed K'}}$ is the total width of the intermediate state.

The integrated cross section can easily be derived from (1) and is given by

$$\int\sigma_{\rm ind}(\omega_{\rm X})\,{\rm d}\omega_{\rm X}\simeq\frac{\lambda_{\rm X_r}^2}{4}\,\Gamma^{\rm rad}(\omega_{\rm X_r};\,{\rm IS}\to\text{`mixed K'})\times\frac{\Gamma^{\rm rad+conv}(\omega_\gamma;\,\text{`mixed K'}\to{\rm F})}{\Gamma^{\rm tot}_{\text{`mixed K'}}}\,, \tag{2}$$

where the energy of the resonant photons is already fixed by the condition $\omega_{\rm X_r}=E_{\text{`mixed K'}}-E_{\rm IS}$.

The cross section of the induced decay of ^{178m2}Hf measured by the Texas Collaboration and reported in Refs [2, 3, 7] proved to be anomalously large. The researchers believe that the spectrum of the excited ^{178}Hf nucleus contains a 'mixed K' level (see Fig. 1) involved in the induced decay. In

all experiments of Collins's group, this intermediate state allowed effectively overcoming the K-forbidding.

The following remark is in order. As the measurement technique becomes more sophisticated and the sensitivity of the measuring devices grows, the decay scheme becomes more involved. From time to time, new, usually low-intensity transitions and sparsely populated levels are added to the scheme. But Refs [2–10] deal with an entirely different case. As we see shortly, the properties that a 'mixed K' state must exhibit in order to comply with the requirements of measurements [2, 3, 7, 10] are unusual, to say the least. Levels with such properties will dramatically change the known decay schemes and the gamma spectra of the nuclei. Today, there is no justification for the existence of such levels except for the results of a few experiments belonging to the above-mentioned group.

Nevertheless, let us adopt all the assumptions put forward in Refs [2–10]. We now consider a model problem with an additional level whose properties comply with the following strong assumptions [22, 23].

Model. 1. We assume that the intermediate 'mixed K' level is connected to all other nuclear states by K-allowed transitions.

2. To this 'mixed K' level, we assign spin 15^-, i.e., we ensure an E1-transition between this level and the isomer 16^+(1446 keV).

3. We assume that the nuclear matrix element of the E1-transition has the largest possible value for the ^{178}Hf nucleus, i.e., the matrix element of the collective transition to a giant-dipole-resonance (GDR) state.

4. A similar assumption concerning the collective nature is made for all transitions from the 'mixed K' state to lower levels. (This assumption reduces the probability of the nucleus returning to the isomer level and creates conditions for maximum population of the states from which enhanced-intensity transitions were recorded in Refs [2, 3, 7, 10].)

Within the adopted model, the value of the radiation width for the E1-transition linking the isomer to the intermediate level located at a distance of 40 keV is

$$\Gamma^{\mathrm{rad}}(\mathrm{E1_{GDR}};\mathrm{IS}\rightarrow\text{'mixed K'};40\ \mathrm{keV})\simeq 3\times 10^{-3}\ \mathrm{eV}\,.$$

To detect an act of induced decay, we must ensure that the nucleus does not return with probability one from the intermediate state to the isomer state. The calculations done in Refs [22, 23] show that populating the nearest state 13^-(2433 keV) is the optimal way of inducing decay in the isomer 16^+(2466 keV). Such population ensures an E2-transition with the minimum possible multipolarity from the intermediate 15^- level with the radiation width $\Gamma^{\mathrm{rad}}(\mathrm{E2_{GQR}};\omega_\gamma=53\ \mathrm{keV})\simeq 10^{-8}$ eV and the conversion coefficient $\alpha\simeq 60$. (The subscript GQR in the expression for Γ^{rad} is an indication that, in accordance with assumption 4 of the model, we use the matrix element of the transition to a giant-quadrupole-resonance state.) The decay of the intermediate state into the 12^-(2136 keV) and 11^-(1859 keV) levels along the radiative and conversion channels adds less than 1% to the probability of transition to level 13^-(2433 keV).

Summarizing, we obtain the following upper bound on the integrated cross section:

$$\int\sigma_{\mathrm{ind}}(\omega_{\mathrm{X}})\,\mathrm{d}\omega_{\mathrm{X}}\leqslant 10^{-27}\ \mathrm{cm^2\ keV}\,.$$

This is roughly a million times smaller than the value measured by Collins et al. [2] ($\sim 10^{-21}$ cm^2 keV). The only way to compensate for such a huge difference is to assume that there is an entire 'continuum', i.e., approximately 10^6 nonoverlapping intermediate levels, located 40 ± 20 keV above the isomer state and exhibiting the remarkable properties 1–4.

Clearly, Eqn (2) contains the possibility for increasing the theoretical value of the cross section. In addition to the above four assumptions, we make one more assumption.

5. Let the spectrum of ^{178}Hf have a level below the intermediate 'mixed K' state and let this level be such that the partial width of the transition to it practically exhausts the total width $\Gamma^{\mathrm{tot}}_{\text{'mixed K'}}$ of the intermediate state. (This hypothesis, as is to be explained in Section 2.2, is implied in Ref. [7].) Setting

$$\frac{\Gamma^{\mathrm{rad+conv}}(\omega_\gamma;\text{'mixed K'}\rightarrow\mathrm{F})}{\Gamma^{\mathrm{tot}}_{\text{'mixed K'}}}\sim 1 \qquad (3)$$

in Eqn (2), we obtain the following estimate for the integral cross section:

$$\int\sigma_{\mathrm{ind}}(\omega_{\mathrm{X}})\,\mathrm{d}\omega_{\mathrm{X}}\leqslant 10^{-23}\ \mathrm{cm^2\ keV}\,.$$

Clearly, the result is roughly 100 times smaller than the measured quantity.

In Ref. [3], the value of the integral cross section of the induced decay through an intermediate 'mixed K' level that is located at a 'distance' no greater than 20 keV from the isomer level is 2.2×10^{-22} cm^2 keV. The theoretical estimate of the cross section for such a level also changes. The radiation width of the E1-transition is proportional to the third power of the energy. Hence, by replacing 40 keV with 20 keV, we obtain

$$\int\sigma_{\mathrm{ind}}(\omega_{\mathrm{X}})\,\mathrm{d}\omega_{\mathrm{X}}\leqslant 10^{-28}\ \mathrm{cm^2\ keV}$$

with assumptions 1–4, and

$$\int\sigma_{\mathrm{ind}}(\omega_{\mathrm{X}})\,\mathrm{d}\omega_{\mathrm{X}}\leqslant 10^{-24}\ \mathrm{cm^2\ keV}$$

with the additional assumption 5. We still need, respectively, six and two orders of magnitude to obtain the measured value.

2.2 Decay in the presence of photon–atomic shell interaction

We now analyze Ref. [7]. In that paper, the Texas Collaboration reports the detection of a 1–3% acceleration of spontaneous decay of the isomer when the L-shell of the hafnium atom is ionized by synchrotron radiation with the photon flux $\varphi\simeq 10^{11}$ photons cm^{-2} s^{-1}. The measured enhancement $f\simeq 0.01-0.03$ makes it possible to calculate the induced decay cross section:

$$\sigma_{\mathrm{ind}}=\frac{f\ln 2}{T^{\mathrm{IS}}_{1/2}\varphi}\simeq 2\times 10^{-22}\ \mathrm{cm^2}\,.$$

The researchers then assumed that the observed effect is caused by the excitation of the ^{178}Hf nucleus from the isomer

state to the intermediate 'mixed K' state level because of the transition of one of the atomic electrons to a vacancy in the L-shell. Knowing the photoionization cross section $\sigma_{\rm ion}^{\rm (L)} \simeq 7.5 \times 10^{-20}$ cm^2 [24], they estimated the probability P of this process as

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

We note that this formula contains no branching factor, which is the same as setting this factor to unity. This, as we have seen, is possible only if there is a level with properties reflected in assumption 5 of the model, located below the intermediate 'mixed K' state.

Now we can analyze the value $\sigma_{\rm ind} \simeq 2 \times 10^{-22}$ cm^2 measured in the experiment described in Ref. [7], first within the framework of assumptions 1–4 and then with allowance for assumption 5.

We assume once more that the excitation of the intermediate 'mixed K' state occurs in the E1-transition of an electron from the M_{II}-shell to the L_I-shell of the hafnium atom (the chosen subshells guarantee that the atomic matrix element is at its maximum [25]). We note that here we are speaking of a 'mixed K' level that is located approximately 8.9 keV above the isomer state 16^+(2446 keV). Within the framework of the process being discussed, the assumption about the resonant nature of the excitation is not as trivial as it is in the case of bremsstrahlung photoexcitation. The intermediate and isomer levels must now be connected by a transition whose energy $\omega_{\rm N}$ coincides with the energy $\omega_{\rm A}$ of an atomic transition within the vacancy width $\Gamma_{\rm L_I} + \Gamma_{\rm M_{II}} \simeq 10$ eV [26]. We thus add an extremely strong assumption to the model in Section 2.1 and call it assumption 6.

The theory of nuclear excitation by electron transition, or NEET, developed in Refs [27–30] is in good agreement with modern experimental data [31–34] and can be used to analyze the results of the Texas Collaboration [7].

The relative probability of the state of the process depicted in Fig. 3, in which the nucleus is excited by a virtual photon emitted by an atomic electron, can be calculated as [27–29]

$$P_{\rm NEET} = \left(1 + \frac{\Gamma_{\rm M_{II}}}{\Gamma_{\rm L_I}}\right) \frac{E_{\rm int}^2}{(\omega_{\rm N} - \omega_{\rm A})^2 + (\Gamma_{\rm L_I} + \Gamma_{\rm M_{II}})^2/4} . \quad (4)$$

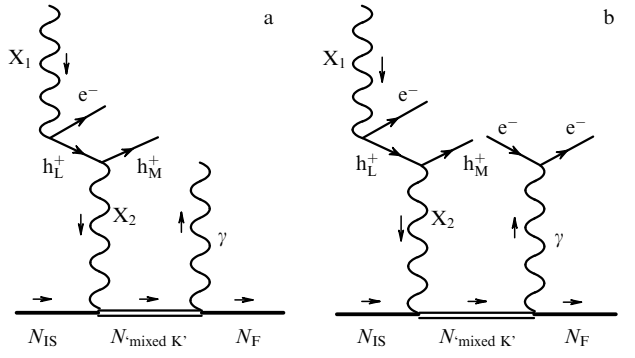

Figure 3. Diagrams of the induced decay with atomic shell ionization. Depicted is the nucleus excitation process in the transition of a formed vacancy from the lower atomic levels to the higher ones followed by the decay of the intermediate nuclear state along the same channels as in Fig. 2.

The interaction energy $E_{\rm int}$ of the electron electromagnetic transition current

$$j^{\mu}_{\rm L_I M_{II}}(\mathbf{r}) = -e\bar{\psi}_{\rm L_I}(\mathbf{r})\gamma^{\mu}\psi_{\rm M_{II}}(\mathbf{r})$$

and nucleus electromagnetic transition current

$$J^{\nu}_{\rm `mixed\ K',\,IS}(\mathbf{R}) = e\Psi^{+}_{\rm `mixed\ K'}(\mathbf{R})\hat{J}^{\nu}\Psi_{\rm IS}(\mathbf{R})$$

is the key parameter in (4), and $E_{\rm int}^2$ is the square of the absolute value of the interaction Hamiltonian

$$H_{\rm int} = \int {\rm d}^3 r\, {\rm d}^3 R\, j^{\mu}_{\rm L_I M_{II}}(\mathbf{r}) D_{\mu\nu}(\omega_{\rm N}; \mathbf{r} - \mathbf{R})\, J^{\nu}_{\rm `mixed\ K',\,IS}(\mathbf{R}) \quad (5)$$

averaged over the initial states and summed over the final states, with $D_{\mu\nu}(\omega_{\rm N}; \mathbf{r} - \mathbf{R})$ being the photon propagator. Numerical calculations for a transition that satisfies assumptions 1 and 2 of the model yield $E_{\rm int} \simeq 1.3 \times 10^{-1}$ eV. This is the maximum possible value of the energy of the interaction triggering the nuclear IS $\rightarrow$ 'mixed K' transition in the electron transition to the L-shell.

With conditions 1–4 and 6 satisfied, the probability $P_{\rm NEET}$ amounts to roughly 1.1×10^{-3}. The branching factor $\beta_{\rm `mixed\ K'}$ in the same model has been calculated in Ref. [25], and the result is $\beta_{\rm `mixed\ K'} < 10^{-3}$. Hence, the upper bound on the induced decay cross section satisfies the relation

$$\sigma_{\rm ind} < 10^{-25}\ {\rm cm}^2 ,$$

i.e., is at least a thousand times smaller than the value measured by the Texas Collaboration [7].

When assumption 5 is taken into account, the upper bound remains four to five times smaller than the experimentally measured value $(1.77-1.95) \times 10^{-22}$ cm^2. It must be noted that the probability of simultaneous implementation of all six assumption is extremely low, not to mention the fact that each assumption by itself appears to be highly improbable.

2.3 Decay in an inverse NEET process

Finally, we check the last hypothesis, namely, that the lifetime of the isomer 16(2446 keV, 31 yr) decreases due to an increase in the probability of an E3-transition with the energy 12.7 keV to the level 13^-(2433 keV). In ordinary conditions, this nuclear transition is the main process in the decay of the isomer (the branching factor $\beta_{\rm E3} = 0.9982$ [12]). Acceleration may be caused by an inverse NEET process involving the atomic shell (Fig. 4) [35]. This is possible, for instance, in the decay of isomer levels in ^{197}Au, ^{193}Ir [35], and other nuclei, provided that the outer atomic shell participating in NEET is ionized. At resonance, this mechanism yields a maximum possible enhancement compared to all other processes involving atomic shells [35].

To obtain the upper bound on the cross section of the process, we make certain assumptions (a customary feature of this report) that ensure optimum conditions for the decay along the channel in question. We suppose that after the L-shell of the hafnium atom has been photoionized, ionization of the outer shells of the atom (an Auger process) occurs in the M $\rightarrow$ L transition with probability one, in which the distance between the states L_{III} and M_{IV} becomes exactly equal to the energy of the nuclear E3-transition 16^+(2446 keV, 31 yr) $\rightarrow$ 13^-(2433 keV). We also assume that the L_{III}-shell is full and the M_{IV}-shell is vacant. This ensures that the inverse NEET (INEET) process can run. The probability of this process occurring can be calculated by the formula

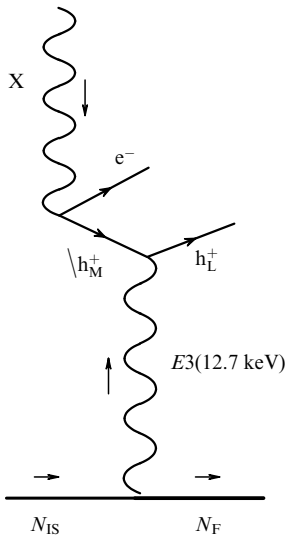

Figure 4. Diagram describing the acceleration of the decay of ^{178m2}Hf in an inverse NEET process.

$W_{\mathrm{INEET}} = \Gamma_{\mathrm{M_{IV}}} P_{\mathrm{INEET}}$ [35], where P_{INEET} is the relative probability of the atom being excited from the $\mathrm{L_{III}}$ state to the $\mathrm{M_{IV}}$ state in the nuclear transition in question. Now this probability can be estimated by formula (4) with the proper initial and final atomic and nuclear states. The interaction energy $E^2_{\mathrm{int}}(\mathrm{E3}; \mathrm{M_{IV}} \to \mathrm{L_{III}}; \mathrm{IS} \to 13^-)$ needed for the INEET process amounts to 1.3×10^{-23} eV^2. This very small value is the result of the nuclear E3-transition being K-forbidden (the reduced probability of the E3-transition in Weisskopf units, $\mathrm{B_{W.u.}(E3)}$, amounts to only 9×10^{-10} [12]). As a result, the INEET relative probability is also very small: $P_{\mathrm{INEET}} \simeq 6 \times 10^{-24}$.

The maximum number of isomeric nuclei that can decay along the INEET channel per unit time, Q_{INEET}, in the Texas Collaboration's experiments [7] under the above conditions can be estimated by the formula

$$Q_{\mathrm{INEET}} \simeq N_{\mathrm{IS}} P_{\mathrm{INEET}} \sigma^{(\mathrm{L})}_{\mathrm{ion}} \varphi_{\mathrm{X}} \,,$$

where N_{IS} is the number of hafnium isomeric nuclei in the target. Comparison of this estimate with the natural decay activity of the isomer, $Q = \lambda_{\mathrm{IS}} N_{\mathrm{IS}}$ (where $\lambda_{\mathrm{IS}} = \ln 2 / T^{\mathrm{IS}}_{1/2}$), yields

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

This result makes it totally impossible to explain the experimental results reported in [7] by the enhancement of the nuclear E3-transition 16^+(2446 keV, 31 yr) $\to$ 13^-(2433 keV) due to the involvement of the atomic shell. There is no way here in which one can use assumptions concerning the nature of the nuclear transition in question (similar to assumptions 1–4 of the model). The intensity of this transition has been measured and no place remains for speculation.

3. From 'mixed K' to 'mixed J'?

In the experiment with a beam of synchrotron radiation [10], where the isomeric nuclei ^{178m2}Hf were irradiated by photons with the energy 9567 eV (which is 6 eV higher than the binding energy of an electron on the $2p_{3/2}$ shell of the hafnium atom, $E_{\mathrm{L_{III}}} = 9561$ eV [36]), a new gamma line with the energy 2457.20(22) keV was detected (see Fig. 1).

In view of what was said in Sections 2.1 and 2.2, this result appears to be highly improbable. The induced decay in the experiment described in Ref. [10] occurred in approximately the same conditions as the one described in Ref. [7]. (No value of the cross section was given in Ref. [10], but the figure for the gamma emission is roughly the same as the one reported in Ref. [7], while the synchrotron-radiation flux is more than 30 times higher: 2×10^{12} cm^{-2} s^{-1} in the 1 eV range against 3×10^{10} cm^{-2} s^{-1} in the 0.5 eV range.) As usual, for effective excitation, the spin of the intermediate state must be $\mathrm{J}^\pi = 15^-$. The multipolarity of the gamma radiation, which in Ref. [10] is interpreted as the result of the decay of the intermediate state into the ground state 0^+ of the ^{178}Hf nucleus, must then be E15. The ratio of the radius R of the nucleus to the wavelength λ_γ is small: $R/\lambda_\gamma = 0.013$. Hence, the probability of the E15-transition, e.g., in the Weisskopf model amounts to only 2.5×10^{-49} s^{-1}.

The equation describing the excitation and decay of the 'mixed K' state in an irradiated target containing N_{m2} ^{178m2}Hf nuclei has the simple form (where we ignore the natural decay of ^{178m2}Hf nuclei)

$$\frac{\mathrm{d}N_{\text{'mixed K'}}}{\mathrm{d}t} = N_{m2}\sigma\varphi - (\lambda + \lambda_{\mathrm{E15}}) N_{\text{'mixed K'}} \,,$$

where λ is the decay constant, which ensures the observed enhancement of ordinary gamma transitions in Refs [2–9], and λ_{E15} is the probability of the emission of an E15(2457.2 keV) gamma quantum in Ref. [10]. The activity of the target along the indicated second channel is

$$Q_{\mathrm{E15}} \simeq N_{m2}\sigma\varphi \, \frac{\lambda_{\mathrm{E15}}}{\lambda + \lambda_{\mathrm{E15}}} \,.$$

We take the value of the cross section of excitation of the intermediate state obtained in the first experiment of Collins's group involving a beam of synchrotron radiation [7]: $\sigma \simeq 2 \times 10^{-22}$ cm^2. We estimate λ in accordance with assumption 5 of the model, because its value must be comparable to the width of the IS $\to$ 'mixed K' transition (see Section 2.1). In this case, it will take 10^{50} years (!) of continuous irradiation of a target with about 10^{13} ^{178m2}Hf isomers in an experiment similar to the one described in Ref. [10] for at least one transition to occur from the intermediate state to the ground state of the nucleus accompanied by emission of an E15(2457.2 keV) photon. The result speaks for itself. Actually, in such a situation, it is irrelevant what multipolarity the transition has, E15 or, e.g., E13 (in the latter case, the cross section of excitation of the intermediate level is much smaller). It is also irrelevant what the synchrotron-radiation flux was or how many ^{178m2}Hf nuclei the target actually contained. Modern physical theories provide no explanation for this experimentally observed fact of the given transition.

Possibly the simplest way to remove this contradiction is to pronounce the intermediate level a 'mixed J' level and to assign to it all the properties that the authors of Refs [2–10] assigned to 'mixed K' states.

4. Induced decay involving known levels

The experimentally induced decay of the isomer ^{178m2}Hf can be implemented according to the diagrams in Fig. 2 by

involving the known nuclear states that are close to the 16^+(2446 keV) level [37]. The existing data (see Ref. [12]) are sufficient for calculating the cross section of the process involving the 14^-(2573 keV) level depicted in Fig. 1. This state splits into three low-lying levels, one of which is our isomer level and the other two are the 13^-(2433 keV) and 12^-(2136 keV) bands $K^\pi = 8_1^-$. The intensities and internal electron conversion coefficients of the indicated transition have been measured and can be found in Ref. [12]. Using these intensities and conversion coefficients, one can easily calculate the width of the radiative M2-transition, $\Gamma_{M2}^{rad}(16^+ \to 14^-) = 1.8 \times 10^{-13}$ eV, and the branching factor for the decay of the 14^-(2573 keV) level into the states 13^-(2433 keV) and 12^-(2136 keV), $\beta = 0.476$. An estimate by formula (2) leads to the following result for the integrated cross section of the induced decay of ^{178m2}Hf involving the 14^-(2573 keV) level:

$$\int \sigma_{ind}(\omega_X)\, d\omega_X \simeq 2 \times 10^{-35} \text{ cm}^2 \text{ keV}.$$

The value obtained by Carroll [37] is close to this value. Such a cross section imposes stringent restrictions on the experiment. For instance, with the synchrotron-radiation beam of the Argonne National Laboratory irradiating a 0.5-by-0.5 cm target that contains 10^{15} isomeric nuclei whose natural activity is 0.7×10^6 s^{-1}, an additional 10^{-6} isomers will decay every second as a result of the induced transition involving the 14^-(2573 keV) level. With a relative increase in the decay activity by a quantity of the order 10^{-12}, even the appearance in the emission spectrum of new lines from the transitions 14^-(2573 keV) $\to$ 13^-(2433 keV), 12^-(2136 keV) simplifies the problem of detecting the effect only slightly.

5. Isomer accumulation

The targets that have been used in induced-decay experiments contain 10^{13} to 10^{16} ^{178m2}Hf nuclei. Some of the methods of production, chemical isolation, and mass separation of such numbers of isomers have been described in Refs [17, 38]. The cross sections of some of the processes are also known. For instance, for the reaction ^{176}Yb$(\alpha, 2n)^{178m2}$Hf, the cross section is $\sigma \simeq 7 \times 10^{-27}$ cm^2 at $E_\alpha = 36$ MeV [39]. In the event of spallation of ^{186}W by 650 MeV protons, the ^{178m2}Hf production cross section is roughly 5×10^{-28} cm^2, while in the fragmentation of the target containing a natural mixture of isotopes of Ta, $\sigma \simeq 3 \times 10^{-28}$ cm^2 [40]. Despite the relatively large cross sections, commercial production of ^{178m2}Hf isomers, e.g., several grams per annum, cannot be achieved by these methods, because there are no devices capable of generating a current of protons or alpha particles sufficiently high for operating with large amounts of the initial material.

Very large targets can be irradiated in power-generating thermal reactors, where a large neutron flux is maintained in the entire volume. However, the cross section of the reaction ^{177}Hf$(n, \gamma)^{178m2}$Hf is small: $\sigma \simeq 2 \times 10^{-31}$ cm^2 [41]. Hence, in one year, such a reactor with an average flux of thermal neutrons of 2×10^{14} cm^{-2} s^{-1} will produce approximately 1 μg of ^{178m2}Hf from 1 kg of the natural mixture of hafnium isotopes (this mixture contains 18.6% of the isotope ^{177}Hf).

Progress in this area might be possible with fusion reactors of the future. The cross section of the process ^{179}Hf$(n, 2n)^{178m2}$Hf is roughly 7×10^{-27} cm^2 for neutrons with an energy of 14 to 15 MeV [42]. If such reactors are built and if a fraction of the neutrons produced in the d + t reaction is used to irradiate ^{179}Hf, we can hope that macroscopic amounts of ^{178m2}Hf will be produced.

The various mechanisms of ^{178m2}Hf production are studied in greater detail in Ref. [43], where, in particular, the ratios σ_{m2}/σ_g of the cross sections of the production of the isomer ^{178m2}Hf and the ^{178}Hf nucleus in the ground state caused by photons with energies amounting to 22 and 4500 MeV on ^{179}Hf and ^{181}Ta targets, respectively, are listed. The reaction ^{179}Hf$(\gamma, n)^{178m2}$Hf at $E_\gamma \simeq 14$ MeV (the GDR region) and higher is of little use for the production of large amounts of the isomer. The neutron must carry away too large an angular momentum, and hence the cross section proves to be relatively small. One obstacle in the way of using photons with energies of several gigaelectronvolts is the difficulty in generating them in quantities needed for commercial production of the isomer.

Finally, the use of nuclei that are in long-lived states with high spin, e.g., ^{180}Ta$(9^-, 77.1$ keV, $> 1.2 \times 10^{15}$ yr), as targets does not help to solve the problem. In this case, all the difficulties in accumulating ^{178m2}Hf are shifted to the production of large quantities of the nuclei in the above states.

6. Conclusion

The production of several grams or more of the isomer ^{178m2}Hf is an extremely difficult task. So far, no effective process for such production has been described in the literature. The use of only one of the above reactions will require large investments. Actually, such burdensome expenditures from state budgets may prove useless: no energy can be liberated by the method described in Refs [2, 3, 7, 10]. The cross sections of the induced decay of the isomer ^{178m2}Hf measured by that method do not agree with the current ideas about the physics of the nucleus and the physics of electromagnetic nuclear processes.

Summarizing the results in the present report, I would like to note the following. Theoretical calculations and the analysis of the existing experimental data suggest that the hafnium problem, as presented by the works of Collins's group, does not exist. The hullabaloo over the hafnium bomb was due to meaningless experimental data and the incompetence of certain individuals rather than to the real possibility of building a radically new weapon based on ^{178}Hf.

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