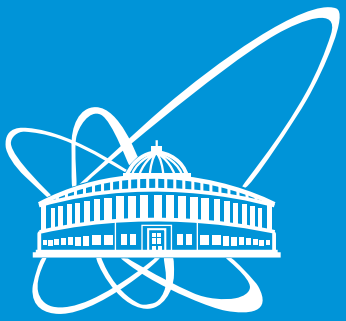

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^{178m2}Hf AND OTHER ISOMERS CANDIDATES FOR THE DECAY STIMULATED BY X-RAY PHOTONS

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1. Introduction

Experiments with high-spin, long-lived nuclear isomers, like ^{180m}Ta and ^{178m2}Hf, are recognized during the decade as an innovating test of the structure of quasiparticle nuclear states and their interaction with external radiation. Triggering of the ^{180m}Ta and ^{178m2}Hf decay via excitation of K-mixed states was observed in experiments on bremsstrahlung irradiations of the isomeric sources. Induced release of the stored nuclear energy is promising for applications such as creation of the pulsed gamma-ray source controlled by soft x-ray device.

The triggered release of isomer energy is very attractive for the production of sources emitting powerful pulses of radiation in the gamma-ray range. For example, self-stimulation of an isomeric transition would create a gamma-ray laser and such a device might also be possible using a triggered release for a precursive step to a lasing transition. There are, however, many problems to be solved since significant barriers are already visible in both of the currently proposed variants for realizing a gamma-ray laser based on a solid-state Mössbauer system [1] and a cooled atomic beam [2]. These difficulties can be relaxed if an easier goal is chosen, namely, the development of a system for generation of γ-ray flashes controlled by a soft x-ray device. Such flashes are not yet laser radiation since they are incoherent and isotropic in space, but they can still be quite powerful and can be useful for many important applications. For instance, suppose 1 mg of the 31-year lived ^{178}Hf^{m2} isomer material was triggered by an x-ray pulse and all nuclei release their energy, stored in the form of excitation energy of the isomeric level, during 1 μs. This would result in the emission of a 1-TW γ-radiation flash. The total energy in the flash might be as high as 1 MJ.

Presently, there are numerous critical questions, such as the nature of the physical mechanism responsible for triggering, that must be addressed before any application can be considered realistically. These questions define the most important areas in which research must concentrate:

1. Analysis of the general properties of isomers and a choice of the best one(s),
2. Development of methods for the production of isomers in mg amounts,
3. Measurements and theoretical descriptions of the integrated cross sections for isomer triggering by x-ray photons, and
4. Proposals for chain-reaction triggering in an ensemble of isomeric nuclei.

In present talk the first 2 areas are mostly of interest. In addition to discussed in literature ^{178m2}Hf and ^{179m2}Hf, the ^{177m}Lu and ^{242m}Am isomers can be identified as the best candidates.

In the context of problem 2 we discuss the production cross-sections for Hf and Lu isomers in the irradiations of the Ta targets by protons at 660, 200 and 100 MeV. They are compared with that ones for the background radioisotopes, and the conclusion is drawn that the irradiations at 100 MeV can be used as more economic way for the

isomer production than the irradiations at Los Alamos 800 MeV high-power beam. At the same time, it is clear that the highest productivity of any radionuclide can be reached using (n,γ) reaction at standard or high-flux reactors. Fortunately, ^{177m}Lu and ^{242m}Am have high enough production cross-section in (n,γ)-reactions.

The third area is typically viewed as the most important at this time, and, in general, there are several different methods that are conceptually possible for the detection of events in which a release of isomer energy is triggered:

1. Detection of short-lived products following triggering of a stable isomer (so-called activation method), for example, as in experiments with ^{180}Ta^{m} [3-6],
2. Detection under the beam of an intensity enhancement in spontaneous decay radiation caused by external irradiation, as in Ref. [7] for ^{178}Hf^{m2},
3. Detection of a new gamma line corresponding to the triggering process that doesn't appear in the spontaneous decay of the isomer,
4. Detection of a unique gamma-ray cascade following a triggering event that is significantly different from the spontaneous-decay cascade,
5. Detection of a loss in isomer activity as a result of 'burning' in an intense irradiation, and
6. Detection of an intensity enhancement in the daughter decay radiation after "burning" the isomer to the radioactive state.

Only the first two methods have been applied so far in experiments and it is proposed here to use third and sixth schemes for the reliable detection of triggering of the ^{242m}Am and ^{177m}Lu isomeric samples, respectively.

2. Properties of isomers

Known isomers with half-lives $T_{1/2} \geq 3$ d are listed in Table 1. Shorter lifetimes are judged to be inconvenient for accumulation of the isomeric material in an amount needed for the triggering experiment.

To select the best candidates, all isomers are estimated by two parameters for ranking: the excitation energy of the isomeric state and the production cross-section in (n,γ) reaction with thermal or resonant neutrons. They define the capability to store as much as possible amount of energy in the isomeric sample. These parameters are reduced in Table 1 following to Refs. [8-10]. The highest excitation energy is observed at the cases of ^{178m2}Hf and ^{179m2}Hf isomers, but their production cross-sections in (n,γ)-reactions are low (unknown) when stable isotopes of Hf are used as targets. The ^{177m}Lu isomer has 2.5 times lower excitation energy than ^{178m2}Hf, but it can be produced in a quantity order of magnitude larger, than ^{178m2}Hf, since (n,γ)-reaction is a producer of the ^{177m}Lu isomeric state. The excitation energy of ^{242m}Am is relatively low, but its' production cross-section is extraordinary high, and it appears to be stored in any reactor as by-product, and can be commercially supplied by the radioisotope producing companies. Finally, four isomers of: ^{178m2}Hf, ^{179m2}Hf, ^{177m}Lu and ^{242m}Am, are selected as the best ones and their nuclear properties are compared below.

Table 1. Nuclear isomer properties and production cross-sections in (n,γ) reactions

Isomer	$T_{1/2}$	I^{π}	E^*, keV	Producer reaction	σ_{th}, barn	I_γ, barn
^{91m}Nb	61 d	$1/2^-$	105	^{93}Nb(γ, 2n)		
^{93}Nb	16.1 y	$1/2^-$	31	^{93}Nb(γ, γ′)		
^{97m}Tc	90 d	$1/2^-$	97	^{96}Ru(n, γ)^{97}Ru ↓ ^{97m}Tc	0.29 10^{-4}(eff.)	7.3 $2\cdot10^{-3}$ (eff.)
^{102m}Rh	2.9 y	6^+	141	^{103}Rh(γ, n)		
^{108m}Ag	418 y	6^+	109	^{107}Ag(n, γ)	0.33	1.2
^{110m}Ag	250 d	6^+	118	^{109}Ag(n, γ)	4.9	72
^{113m}Cd	14.1 y	$11/2^-$	264	^{112}Cd(n, γ)	<2	-
^{114m1}In	49.5 d	5^+	190	^{113}In(n, γ)	8.1	220
^{117m}Sn	13.6 d	$11/2^-$	315	^{116}Sn(n, γ)	0.006	0.49
^{119m}Sn	293 d	$11/2^-$	90	^{118}Sn(n, γ)	0.01	-
^{121m}Sn	55 y	$11/2^-$	6.3	^{120}Sn(n, γ)	0.001	-
^{121m}Te	154 d	$11/2^-$	294	^{120}Te(n, γ)	0.34	-
^{123m}Te	119.7 d	$11/2^-$	248	^{122}Te(n, γ)	0.44	5.1
^{125m}Te	57.4 d	$11/2^-$	145	^{124}Te(n, γ) ^{124}Sn(n, γ)^{125}Sn ↓ ^{125}Sb ↓ ^{125m}Te	1.1 0.13 10^{-3}(eff.)	1.4 8 0.05 (eff.)
^{127m}Te	109 d	$11/2^-$	88	^{126}Te(n, γ)	0.063	0.64
^{129m}Te	33.6 d	$11/2^-$	106	^{128}Te(n, γ)	0.027	0.21
^{129m}Xe	8.9 d	$11/2^-$	236	^{128}Xe(n, γ)	0.48	38
^{131m}Xe	11.8 d	$11/2^-$	164	^{130}Xe(n, γ)	0.45	16
^{148m}Pm	41.3 d	6^-	138	^{148}Nd(d, 2n)		
^{166m}Ho	1200 y	7^-	6	^{165}Ho(n, γ)	3.5	20
^{174m}Lu	142 d	6^-	171	^{175}Lu(γ, n)		
^{177m}Lu	161 d	$23/2^-$	970	^{176}Lu(n, γ)	2.8	4.7
^{178m2}Hf	31 y	16^+	2446	^{181}Ta(p, α)		
^{179m2}Hf	25 d	$25/2^-$	1106	^{178}Hf(n, γ)	-	-
^{180m}Ta	$>10^{15}$ y	9^-	75	Stable		
^{184m}Re	169 d	8^+	188	^{184}W(d, 2n)		
^{186m}Re	$2\cdot10^5$ y	8^+	149	^{185}Re(n, γ)	0.3	~5
^{192m}Ir	241 y	9	155	^{191}Ir(n, γ)	0.16	~0.5
^{193m}Ir	10.5 y	$11/2^-$	80	^{192}Os(d, n)		
^{193m}Pt	4.33 d	$11/2^-$	150	^{192}Pt(n, γ)	2.2	~20
^{195m}Pt	4.02 d	$11/2^-$	259	^{194}Pt(n, γ)	0.1	3.1
^{242m}Am	152 y	5^-	49	^{241}Am(n, γ)	54	195

The latter two isomers have lifetimes much longer than the ground states, and, correspondingly, the g.s. nuclei are present in the isomeric sample only as an equilibrium daughter product. This makes such sources practically pure isomeric sources, unlike to Hf isomers, which are characterized in detail in Refs. [11,12] and elsewhere.

Excited levels and the decay schemes of isomers can be found in the Handbook of Ref. [8] or in a form of computer data file in Ref. [13]. As known, ^{177m}Lu isomeric state has an excitation energy of 970 keV, and it decays with a half-life of 161 d mostly (78%) to the 1 s lived ^{177m}Hf isomer, and with the probability of 22% to the ground state of ^{177g}Lu via a multistep γ-cascade. ^{177g}Lu has half-life of 6.7 d, and the β^--decay is accompanied by the another γ-cascade.

Very schematically, the decay of ^{177m}Lu is illustrated in Fig. 1. Three different γ-cascades follow the isomeric transition from ^{177m}Lu and the β^--decays of m and g states to the levels of ^{177}Hf, respectively. By the methods of a precise γ-spectroscopy, it would be possible to distinguish the different cascades and differentiate the branches of ^{177m}Lu and daughter nuclei decays.

Let us to discuss now a thinkable experiment on triggering of the ^{177m}Lu isomer. The same as in the case of the experiment on triggering of the ^{178m2}Hf isomer, Ref. [7], one expects, that after excitation of the ^{177m}Lu isomer by x-ray photon the intermediate state should be populated and decayed promptly through a high-multiplicity cascade by the levels of ^{177}Lu. Thus, the γ-lines of the cascade I in Fig. 1 can be enhanced under the x-ray beam. Detection of the enhancement is just an application of the method 2 (in the list given in the Introduction), which was tested in Ref. [7] for ^{178m2}Hf. This method, being applied to ^{177m}Lu, is somewhat disadvantagable, because much shorter half-life of ^{177m}Lu in comparison with ^{178m2}Hf. Indeed, the rate of decay via cascade I is defined by the equation

$$r = 0.22\frac{\ln 2}{T_{1/2}(177m)}, \qquad (1)$$

and numerically, it is larger by a factor of 14 than the rate of spontaneous decay of ^{178m2}Hf ($T_{1/2}$ = 31 y). Consequently, one has to irradiate the sample by 14 times larger x-ray photon flux, in order to reach the same, as for ^{178m2}Hf, relative enhancement of the spontaneous radiation lines. It means the sensitivity of measurements is 14 times lower, provided the same other physical parameters in both cases, for instance, the integrated cross-section value for the excitation of the intermediate state. The cross-section is unknown, and this is a goal of the

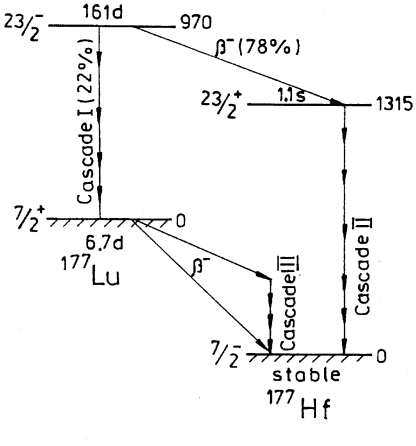

Fig. 1. Fragmentary scheme of the ^{177m}Lu isomer decay.

experiment to estimate this value for ^{177m}Lu.

It would be desirable to improve the sensitivity of the experiment, and the decay properties of ^{177m}Lu allow to apply method 6, see in Introduction. The equilibrium rate of the daughter decay of ^{177g}Lu, followed by cascade III in Fig. 1, should be enhanced after one week irradiation by a factor almost equal to the enhancement of the cascade I under exposure. This is because the triggered depopulation of the isomer leads to an additional production of the ^{177g}Lu state, and this additional yield is accumulated to reach an equilibrium level, the same as in the spontaneous decay of the mother nuclide, ^{177m}Lu. Finally, the enhancement of the cascade III can be measured after exposure in the out-of-beam condition. The latter variant is quite attractive in the sense of experimental conditions. The γ-spectroscopic measurements "in-beam" are stressed much by the background count rate due to the loading of the detector by the direct and scattered photons of the beam. At high intensity irradiations one has to construct sophisticated collimation and shielding systems to isolate the detector from the background radiation. Finally, this increases the distance R between the exposed sample and x-ray source, and the irradiation flux is respectively decreased by a factor proportional to R^{-2}, as known.

Gamma-spectroscopy in "out-of-beam" conditions is much more convenient, and allows to use the orders of magnitude larger photon fluxes when the irradiated sample is placed just near the source of x-rays. Switching off the latter one, all problems of the beam background can be excluded, and no special shielding is needed. Thus, the decay properties of the ^{177m}Lu met the requirement of the higher flux irradiation to reach similar, or even an order of magnitude larger sensitivity in the triggering experiment, than in the known case of the ^{178m2}Hf sample, Ref. [7]. The variant of the daughter-decay detection after an intense x-ray irradiation has to be applied for that (method 6 in Introduction).

After the discussion of technical variants of the triggering experiment with ^{177m}Lu, let us to ask, is it probable that the triggering process can be observed successfully in this case, like it was shown in Refs. [6,7] for ^{180m}Ta and ^{178m2}Hf? The level schemes of these 3 isotopes are different, see the fragments of them in Fig. 2. In a high-sensitivity experiment with the ^{180m}Ta enriched target, Ref. [6], it was shown that first triggering level is placed at an excitation energy of 1070-1080 keV, i.e. about 1000 keV higher than the isomeric state. For ^{178m2}Hf such a level was found in Ref. [7] near 2470 keV, but one have to realize that the ground state of the ^{178}Hf even-even isotope has a decreased mass (increased binding energy) due to the pairing energy. As a result, the ground state is deepened in energy by $2\Delta \approx 1400$ keV (Δ is pairing energy), and for the comparison with odd isotopes, one has to introduce some effective ground state, which is placed higher the g.s. by 1400 keV, as shown in Fig. 2. The triggering level in ^{178}Hf, thus, lies over the effective g.s. by 1070 keV, i.e. essentially at the similar position, like that one in the odd ^{180}Ta. It would be possible to assume, that the level scheme of odd ^{177}Lu also contains the triggering level near an excitation energy of 1070 keV. This is by 100 keV over the isomeric state of ^{177m}Lu, and one can assume a successive triggering of this isomer by soft x-ray photons with an energy of about 100 keV. Such a speculation cannot be qualified as a reliable estimation, and only the

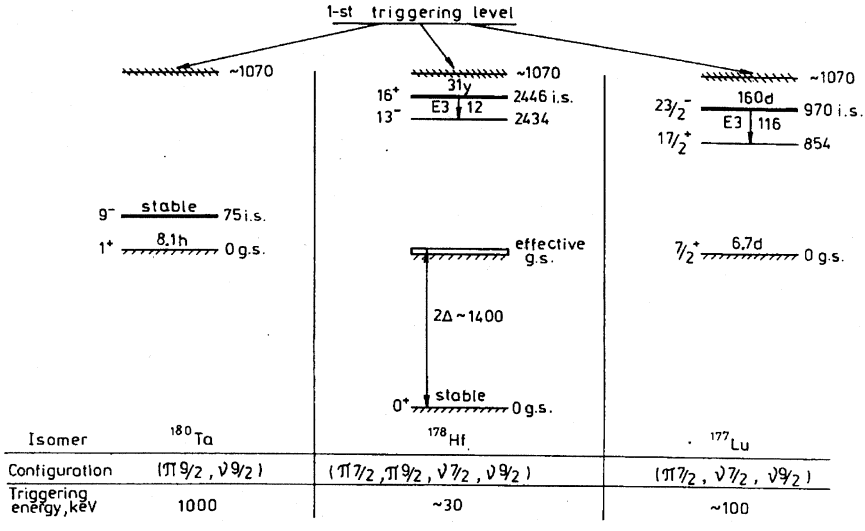

Fig. 2. Positions of the ground, isomeric and triggering levels in ^{180}Ta (Ref. [6]), ^{178}Hf (Ref. [7]), and ^{177}Lu (estimation).

experimental test may prove or disprove the triggering of ^{177m}Lu. This experiment looks important, because ^{177}Lu has advantagable level scheme, high production yield and other convenient properties.

Let us to discuss now the levels of ^{242}Am, they are shown schematically in Fig. 3. The long-lived ($T_{1/2}$ = 141 y) isomeric state has low excitation energy (48.6 keV) and quantum numbers of I, K^{π} = 5, 5$^-$. The triggering level with the K-mixed wave function may be placed at high enough energy, like 1 MeV, if the analogy with ^{180}Ta is valid. But there is known the 3,0$^-$ state at E^* = 52.9 keV, i.e. only by 4 keV above the isomeric level. The 3$^-$ state might be used as an intermediate state if it can be excited by soft x-ray photons when started from the isomeric state. The electric quadrupole transition from 5$^-$ to 3$^-$ level may have very low reduced probability B(E2), because of 3 folded K-hindrance factor, which produces the retardation of the transition by a factor of about 10^3 - 10^4. So, the possibility to use the 3$^-$ level as a mediating state for triggering of the 5$^-$ isomer is not yet clarified, since

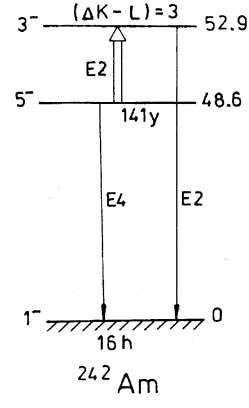

Fig. 3. Levels of ^{242}Am at low excitation energy.

the B(E2) value for $5^- \rightarrow 3^-$ excitation is unknown. It would be important to test experimentally such a transition in ^{242}Am, because, being excited, 3^- state decays promptly to the ground state. Thus, the isomeric energy is released in the short wave-band range of E_γ = 52.9 keV after triggering. The ^{242m}Am isomer is available commercially in an amount of milligrams, or even grams, and a lot of energy can be stored and released if the triggering is successful. This is despite low enough specific energy per one nucleus stored by the ^{242m}Am isomer sample.

Due to the described above reasons, it would be important to carry out the experimental attempt of the ^{242m}Am isomer triggering by soft x-ray radiation. In the band of 4 keV an intense flux of x-rays can be generated using the sources of different principles of action. Among them are the synchrotron and undulator systems, plasma sources and others. A high sensitivity of measurements can be provided by the high intensity of the incident radiation. And let us to discuss now, whether a high sensitivity can be reached also in the detection channel, taking into account the background radiation and the selectivity for the detection of the triggering events.

^{242m}Am decays by the converted isomeric transition to the short-lived ($T_{1/2}$ = 16 h) ground state, and then the radiation of daughter and granddaughter nuclides follows. The characteristic x-ray and α-decay energies of ^{242m}Am, as a precursor, are listed in Table 2. In the γ-spectroscopy experiment, the high-resolution detector (probably Ge detector) can be shielded completely from the penetration of α-particles and conversion electrons. Also, direct or scattered photons from the incident radiation beam are not dangerous, because soft x-ray photons in the range of $E_x \leq 10$ keV can be absorbed by thin enough material, which doesn't disturb the transmission of higher energy photons, namely, 52.9 keV γ-line is of our interest. This line is not emitted in the spontaneous decay of ^{242m}Am, and this situation makes the described experiment, practically, free of a background.

One needs to irradiate ^{242m}Am source by 4-5 keV x-ray beam, and search for the line of 52.9 keV in the spectrum of emitted radiation, which appears only in presence of the beam. This is variant 3 in the list of methods given in the Introduction. Fortunately, all characteristic γ- and x-ray lines of the spontaneous radiation have lower energies, than 52.9 keV, as clear from Table 2. The exception is the K-X rays of Pu emitted with the yield of about 17% in the EC-decay of ^{242g}Am daughter nuclei. Converted γ-transitions create only L-X rays, because the γ-ray energies are not enough for the K-vacancy ionization, but in the EC-decay the characteristic K-X rays are emitted. They penetrate to the detector through all absorbers and generate the Compton continuum at lower energies, including the range near 52.9 keV at the position of the searched for triggering line. This background should be reduced using a high-quality Ge detector, providing the best peak-to-valley ratio. In addition, the background is not differentiated, it is just a continuous spectrum, this is helpful for the subtraction of it.

Finally, one can estimate, that the sensitivity of the triggering experiment with the ^{242m}Am sample might be by 4-5 orders of magnitude higher than at the described in literature case of ^{178m2}Hf triggering, Ref. [7]. It would be difficult to predict now, whether this high sensitivity of the experiment is enough to detect the triggering of 5^-

isomer of ^{242}Am via 3$^-$ state, or not. It depends on the unknown nuclear spectroscopy properties of the transition, and only experiment can clarify the question.

Table 2. Radiation emitted in the decay of ^{242m}Am ($T_{1/2}$ = 141 y)

Gamma radiation		
E_γ, keV	I_γ/100 decay	Origin
LX ray of Pu	10.6	after EC of ^{242}Am^g
	20.6 x k*)	after α decay of ^{242}Cm
LX ray of Am	99.5	after isomeric transition
LX ray of Cm	45.6	after β$^-$ decay of ^{242}Am^g
42.1	0.038	after β$^-$ decay of ^{242}Am^g
44.1	0.027 x k	after α decay of ^{242}Cm
44.5	0.014	after EC of ^{242}Am^g
48.6	about 0.0002	isomeric transition
49.4	0.188	after α decay of ^{242}Am^m
KX ray of Pu	17.3	after EC of ^{242}Am^g
Alpha radiation		
E_α, keV	E_α, keV	Origin
5246	0.41	decay of ^{242}Am^m
5550	24 x k	decay of ^{238}Pu
5593	59 x k	decay of ^{238}Pu
6172	20.6 x k	decay of ^{242}Cm
6216	62.1 x k	decay of ^{242}Cm

*) Coefficient "k" depends on the age of the source. It is of about 1 for ^{242}Cm after 1 year age, and typically <<1 for ^{238}Pu.

3. Production of isomers

Application of nuclear isomers as a reservoir for the energy storage and release requires, as minimum, milligrams of the isomeric material. So that, the high-productivity methods of the isomers accumulation have to be developed, and the possibilities are discussed below.

For high-spin isomers of Hf the reactions with α-particles and heavy-ions have, obviously, the best cross-sections and highest isomer-to-ground state ratios, Refs. [11,14]. But the total yield is limited by a thin target layer, and the restrictions onto the beam intensity. Maximum known quantity (about 10^{17} atoms) of ^{178m2}Hf was produced using spallation reaction in interaction of 800 MeV protons with thick Ta target, as described in Ref. [15]. The massive Ta target irradiated [15] by the 0.4 mA proton beam at Los Alamos meson factory (LAMPF) or the beam dumps of LAMPF accelerator were chemically processed and the isolated Hf fraction contained the ^{178m2}Hf nuclides in a 300 μgs amount, in total. Such a method was not optimized for the best purity and highest yield of the produced ^{178m2}Hf material. Practically, this activity was created as

by-product after operation of LAMPF for other experiments. In addition, an operation of this facility at near-by 1 MW power of the beam is extremely expensive and the irradiated massive targets have extraordinary high total activity. It would be reasonable to propose somewhat more economic method for the production of ^{178m2}Hf, ^{179m2}Hf and other isomers. To test the possibilities of the optimization of the isomer accumulation in proton-induced reactions, we carried out, recently, the irradiations at LNP JINR synchrocyclotron, and detected the radionuclides produced in the Ta target at 100, 200 and 660 MeV proton irradiations. The experiment was sponsored by IGE Foundation, Bucharest, and the results should be published in full volume elsewhere with participation of Drs. J.Adam, V.G.Kalinnikov and group of radiochemists, responsible for the chemical processing of the radioactive targets. Now some preliminary results and conclusions of these studies are discussed.

The absolute yields and production cross-sections are given in Table 3 for many radionuclides from Ta to Cs, including 5 isomers of Hf, Lu and Pm, as they was were measured by the γ-spectroscopy and radiochemistry methods after 660 MeV irradiation. The isomeric-to-ground state ratios were estimated to be higher unity for 6$^-$ isomers of ^{174m}Lu and ^{148m}Pm, while they are much lower for high-spin isomers: to be about 0.1, 0.02 and 0.03 for ^{177m}Lu, ^{178m2}Hf and ^{179m2}Hf, respectively. These values do not contradict to the estimated angular momentum distributions for the products of the reactions induced by 660 MeV protons.

Table 3. Yields of radionuclides and mean cross-sections measured after activation of a 33.3 g/cm^2 thickness Ta target by 660 MeV protons. The energy range is from 660 to 615 MeV for all reactions.

Nuclide	$T_{1/2}$	Type of yield	Yield value *), atoms/proton	Mean σ, mbarn
**) ^{182}Ta	115 d	Indep.	$0.85\cdot10^{-3}$	-
**) ^{181}Hf	42.4 d	Indep.	$1.7\cdot10^{-5}$	0.14
^{179m2}Hf	25.1 d	Indep.	$5.8\cdot10^{-5}$	0.52
^{178}W	21.6 d	Indep.	$6.6\cdot10^{-4}$	5.9
^{178m2}Hf	31 y	Indep.	$3.5\cdot10^{-5}$	0.31
^{177m}Lu	161 d	Indep.	$2.8\cdot10^{-5}$	0.25
^{175}Hf	70 d	EC cum.	$6.2\cdot10^{-3}$	56
^{174g}Lu	3.31 y	Indep.	$1.7\cdot10^{-4}$	1.5
^{174m}Lu	142 d	Indep.	$5.3\cdot10^{-4}$	4.8
^{173}Lu	1.37 y	EC cum.	$7.7\cdot10^{-3}$	70
^{172}Hf	1.87 y	EC cum.	$5.2\cdot10^{-3}$	47
^{172}Lu	6.7 d	Indep.	$0.9\cdot10^{-3}$	8.1
^{171}Lu	8.22 d	EC cum.	$7.8\cdot10^{-3}$	71
^{170}Lu	2.0 d	EC cum.	$7.1\cdot10^{-3}$	64
^{169}Yb	32.0 d	EC cum.	$8.3\cdot10^{-3}$	75

Table 3 (continuation)

Nuclide	$T_{1/2}$	Type of yield	Yield value *), atoms/proton	Mean σ, mbarn
^{168}Tm	93.1 d	Indep.	$1.5 \cdot 10^{-4}$	1.3
^{167}Tm	9.24 d	EC cum.	$7.9 \cdot 10^{-3}$	72
^{166}Dy	3.4 d	β^- cum.	$2.0 \cdot 10^{-4}$	1.8
^{160}Tb	72.3 d	Indep.	$1.2 \cdot 10^{-5}$	0.10
^{156}Tb	5.35 d	Indep.	$5.3 \cdot 10^{-5}$	0.48
^{156}Eu	15.2 d	β^- cum.	$1.3 \cdot 10^{-5}$	0.12
^{149}Gd	9.4 d	EC cum.	$3.0 \cdot 10^{-3}$	27
^{149}Eu	93.1 d	EC cum.	$3.1 \cdot 10^{-3}$	28
^{148}Eu	54.3 d	Indep.	$0.8 \cdot 10^{-4}$	0.73
^{148g}Pm	5.37 d	Indep.	$5 \cdot 10^{-6}$	0.045
^{148m}Pm	41.3 d	Indep.	$7.5 \cdot 10^{-6}$	0.067
^{147}Eu	24.6 d	EC cum.	$1.68 \cdot 10^{-3}$	15
^{146}Gd	48.3 d	EC cum.	$1.54 \cdot 10^{-3}$	14
^{146}Eu	4.6 d	Indep.	$5.4 \cdot 10^{-5}$	0.5
^{145}Eu	5.94 d	EC cum.	$0.94 \cdot 10^{-3}$	8.5
^{144}Pm	363 d	Indep.	$\leq 2 \cdot 10^{-4}$	≤1.7
^{143}Pm	265 d	EC cum.	$6.5 \cdot 10^{-4}$	5.8
^{140}Ba	12.75 d	β^- cum.	$1.2 \cdot 10^{-6}$	0.010
^{136}Cs	13.16 d	Indep.	$\leq 1.3 \cdot 10^{-6}$	≤0.011

*) Random errors are within ±7%, and the standard error of the absolute calibration is of about ±15%;
**) ^{182}Ta and ^{181}Hf are produced in the (n,γ) and (p,pπ$^+$) reactions, respectively.

On the first step of the reaction, colliding proton transfers some linear and angular momenta to the target nucleus. Emission of many nucleons from the excited nucleus leads to the spreading of the angular momentum (l) distribution. Finally, wide l distribution of the reaction residue serves as a precursor to the population of the high-spin isomeric state. The angular momentum distributions at 660 MeV are illustrated in Fig. 4. Mean angular momentum, l_i, transferred in the collision, is shown in Fig. 4a as a function of a number of emitted in the reaction nucleons $(-\Delta A) = A_t + 1 - A_p$, where A_t and A_p are mass numbers of the target and product nuclei. This function has a maximum, because at head-on collisions large linear momentum can be transferred, but the impact parameter is low, and at peripheral collisions the impact parameter is high, but the transferred linear momentum is low. In Fig. 4b the l_i distribution is shown, corresponding to the transfer of linear momentum and excitation energy, which would be enough for emission of 10 nucleons, $(-\Delta A) = 10$. And Fig. 4c shows final distribution after random summation of angular momenta of all 10 nucleons emitted. Such

schematical estimations help us to understand the isomer-to-ground state ratios in the reactions with protons at intermediate energies.

Energy dependences of mean cross-sections are presented in Table 4 for the isomers of interest, and for the background isotopes appeared as contaminants. The ^{172}Hf long-lived ($T_{1/2}$ = 1.87 y) isotope generates an intense γ-radiation background during a time scale of tens years. ^{175}Hf (70 d) decays to the level of 10^{-3} after 2 years cooling, and it is not so disturbing as ^{172}Hf. ^{178}W (21.6 d) is produced with high cross-section, and, after the decay through a short-lived ^{178}Ta, populates the ^{178}Hf ground state. There is a possibility to remove ^{178}W by the methods of radiochemistry, before it is decayed. This operation is necessary in order to conserve the highest isomer-to-ground state ratio for ^{178m2}Hf, as it appears promptly in the reaction without cumulative additions to the yield of ^{178g}Hf.

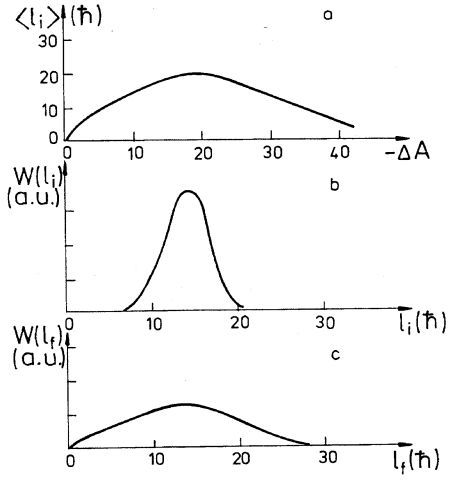

Fig. 4. Results of angular momentum estimations:

a) Mean angular momentum released on the first stage of the reaction as a function of (-ΔA) parameter;
b) Angular momentum distributions for (-ΔA) = 10 after first step of the reaction;
c) The same as in b), but after the nucleon-emission stage.

Table 4. Comparison of the production cross-sections for isomers and for the background radionuclides, as measured at p + Ta reaction

Nuclide	Cross-section, mbarn*)		
	E_p = 660 MeV	E_p = 200 MeV	E_p = 100 MeV
^{179m2}Hf	0.52	0.37	0.29
^{178m2}Hf	0.31	0.30	0.22
^{177m}Lu	0.25	0.12	0.13
^{178}W	5.9	32	150
^{175}Hf	56	140	370
^{172}Hf	47	110	40

*) Random errors are within ±7%.

In Table 4 one can see, that the cross-sections for the isomers remain to be high enough when the proton energy is decreasing from 660 to 100 MeV. Thus, one can propose to use the 100 MeV irradiations as a more economic way for the accumulation of ^{178m2}Hf and ^{179m2}Hf isomers than high power irradiations at Los Alamos accelerator. At 100 MeV the absolute yield of ^{178m2}Hf is lower than at 800 MeV, but it is still as high as of about 10^{10} atoms/sec (2 μg per week), if 150 μA beam is used. Significant advantages of this variant in comparison with Ref. [14] method are visible like these:

1. Two orders of magnitude lower expenses for the operation of a relatively small accelerator supplying a 100 MeV protons. Such kind facilities are constructed in many places. For instance, in Europe exist the accelerators at Uppsala, at Zürich and at Grönningen, which supply the beams of protons with an appropriate energy.
2. Two orders of magnitude lower total activity and much lower variety of produced radioisotopes. This moderates completely the safety problems in a processing of the irradiated targets.
3. (30-50) times lower weight of the target, which is important for the convenience of chemical treatments.

The advantages listed above make, perhaps, a 100 MeV proton irradiation of Ta to be an optimum for ^{178m2}Hf production within the group of reactions induced by charged particles. However as known, charged particles have, in general, low productivity, in comparison with irradiations by slow neutrons in nuclear fission reactors. Unfortunately, the production of ^{178m2}Hf and ^{179m2}Hf in reactors hardly can be effective, because slow neutrons bring not enough angular momentum into a compound nucleus for the population of high-spin isomeric states. Fast neutrons with an energy above the threshold of (n,2n)-reactions are much better in this respect, but it would be difficult to generate the flux of them comparable with the thermal neutron flux in reactors. Thus, the production of Hf isomers remains to be technically difficult problem, and the productivity is restricted.

At the same time ^{177m}Lu and ^{242m}Am are produced in the (n,γ) reaction with high cross-section, and any standard, or high-flux reactor can be used for their production. Table 5 illustrates the advantages of neutron irradiation, when the production of ^{177m}Lu in standard reactor is compared with the highest productivity experiment for ^{178m2}Hf at LAMPF. All parameters are much better in the case of neutron irradiation (^{177m}Lu as a product), namely, much lower weight of the target, higher productivity, much lower total activity and very good isomer-to-ground state ratio. In addition to that, moderate expenses for reactor irradiations should be compared with the extraordinary high at the case of high power LAMPF accelerator. ^{242m}Am is even better than ^{177m}Lu, because it is commercially available being one of the radioisotopes produced in standard reactor operations.

Summary

Experiments on triggered depopulation of nuclear isomers are interesting for nuclear structure studies and important as necessary step on the way to the γ-ray laser creation. Possible schemes for the detection of triggering with high sensitivity are

Table 5. Production of ^{177m}Lu in reactor and ^{178m2}Hf at LAMPF facility (comparison)

Isomer	^{177m}Lu	^{178m2}Hf
Facility	Standard reactor	LAMPF *)
Projectile	Thermal neutrons	Protons, 800 MeV
Flux	10^{14} n/s · cm^2	$2 \cdot 10^{15}$ p/s · 30 cm^2 (0.35 mA)
Target	1 g of ^{176}Lu (enriched)	800 g of Ta (natural)
Productivity	10^{12} at/s	$2 \cdot 10^{11}$ at/s
4 month storage	3.3 mg ($1.1\ 10^{19}$ at.)	0.6 mg ($2 \cdot 10^{18}$ at.)
Target activity after 1 month cool	~10 Ci	~1000 Ci
Method of isolation and typical efficiency	Mass-separation, 30%	Chemistry + mass-separation, ~10%
Isomer-to-ground state ratio	110	~0.002
Cost	Medium	Very high

*) Parameters of irradiation at LAMPF are taken in accordance with Ref. [15], and the productivity of p-induced reaction is measured at Dubna 660 MeV accelerator.

discussed. The long-lived isomer properties are analyzed, and the best candidates are selected. Among them ^{177m}Lu and ^{242m}Am isomers are indicated. The concrete experimental schemes for the observation of triggering process are proposed with account of the individual level schemes and other properties of ^{177m}Lu and ^{242m}Am. The problem of the isomer accumulation in an amount of milligrams is considered. The results on experimental production of Hf and Lu isomers in proton irradiations at intermediate energies are presented and the optimum conditions are discussed. The production of isomers in (n,γ) reaction at standard irradiations in reactors may be much more effective, than in any charged-particles-induced reactions. Fortunately, ^{177m}Lu and ^{242m}Am are among that isomers which can be produced in the (n,γ) reaction with high cross-section.

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^{178m2}Hf и другие изомеры — кандидаты на наблюдение распада, стимулированного рентгеновскими фотонами

Долгоживущие ядерные изомеры возникают благодаря случайной комбинации квантовых чисел начального и конечного состояний изомерного перехода. Правила отбора электромагнитного распада обеспечивают в некоторых случаях высокий фактор запрета, тормозящий распад изомера. В результате известно небольшое число ядерных состояний, живущих месяцы и годы, и нет оснований ожидать открытия большого числа новых. Долгоживущие состояния изучены достаточно хорошо, известны их квантовые числа, схемы распада и иногда — электромагнитные моменты. На сегодня актуальной проблемой является взаимодействие изомерных состояний с внешним излучением, а именно: измерение сечений ядерных реакций, возбуждение вышележащих уровней и поиск проявления структурных особенностей в ядерных реакциях.

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Karamian S.A. E6-2000-233

^{178m2}Hf and Other Isomers Candidates for the Decay Stimulated by x-Ray Photons

Long-lived isomeric states arise randomly, because of the special combination of quantum numbers of initial and final states of the isomeric transitions. Selection rules of the electromagnetic decay provide in some cases high hindrance factors retarding the isomeric decay. As a result, not many nuclear states lived months and years are known, and it is difficult to expect the discovery of a number of new ones. Long-lived states are already studied enough, their quantum numbers, decay schemes and sometimes electromagnetic momenta are known. Relevant problem on today is the interaction of the isomeric states with an external radiation, measurements of the cross-section of nuclear reactions, excitation of higher lying states and searching for manifestation of the structure peculiarities in nuclear reactions.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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