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Author(s): Hsu, Hsiao-Hua
Talbert, Willard L.
Ward, Tom

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THE CREATION AND DESTRUCTION OF $^{178m2}\text{Hf}$ ISOMER by NEUTRON INTERACTIONS

Hsiao-Hua Hsu^{1,2}, Willard L. Talbert², Tom Ward²

1. Los Alamos National Laboratory Los Alamos, NM

2. TechSource, Inc. Los Alamos, NM

INTRODUCTION

The property of the isomer state in $^{178m2}\text{Hf}$ was an interesting topic in nuclear structure studies during the time period 1970 to 1980. The state at 2.446 MeV with spin and parity $K^\pi = 16^+$, has a half-life of 31 years. The isomer is described as a four-quasi-particle state. The K forbidden de-excitation by gamma emission is the reason for long half-life. During 1980, the isomer became a troublesome issue for radiation safety workers, because this isomer can also be produced in the first wall of a fusion reactor containing tungsten and also in a tungsten beam stop of a high-energy accelerator.

In 1985, we were the first¹ to propose several possible ways to induce a fast de-excitation: an incoming particle, such as photon, electron, or nucleon, would change the $^{178m2}\text{Hf}$ nucleus from its isomer level to a different nearby level that has a much shorter lifetime, and to release its energy in the form of gamma-ray de-excitation. We did not perform the experiment at that time, because of the unavailability of adequate $^{178m2}\text{Hf}$ samples.

The debate over the possibility and the mechanism of the so-called ‘triggering’ a fast decay of the isomer state started when Prof. C. B. Collins of The University of Texas published his first article in 1999². In the following decade, many mechanisms have been proposed, and many experiments have been carried out, without any solid conclusions. In Oct. 2008, Hartouni et al from Lawrence Livermore National Laboratory presented a review article³ summarizing the past many experimental results, and interpretations. It listed more than 160 references.

After the x-ray induced de-excitation controversy in 1999, we submitted our second article to APPC 2000⁴. We did further analysis of neutron interaction with the isomer state. The fact that the 16^+ state can be produced by neutron capture of ^{177}Hf indicates that the incoming neutron can induce changes in single-particle orbital. The ground state of ^{177}Hf has a configuration of $\{(7/2[404]_p)^2_0 + 7/2[512]_n\}_{7/2}$. To populate the 16^+ state, the incoming neutron must be in the higher neutron orbital $9/2[624]$ as well as exciting one proton from $7/2[404]$ to $9/2[514]$. (See the following figure 1)

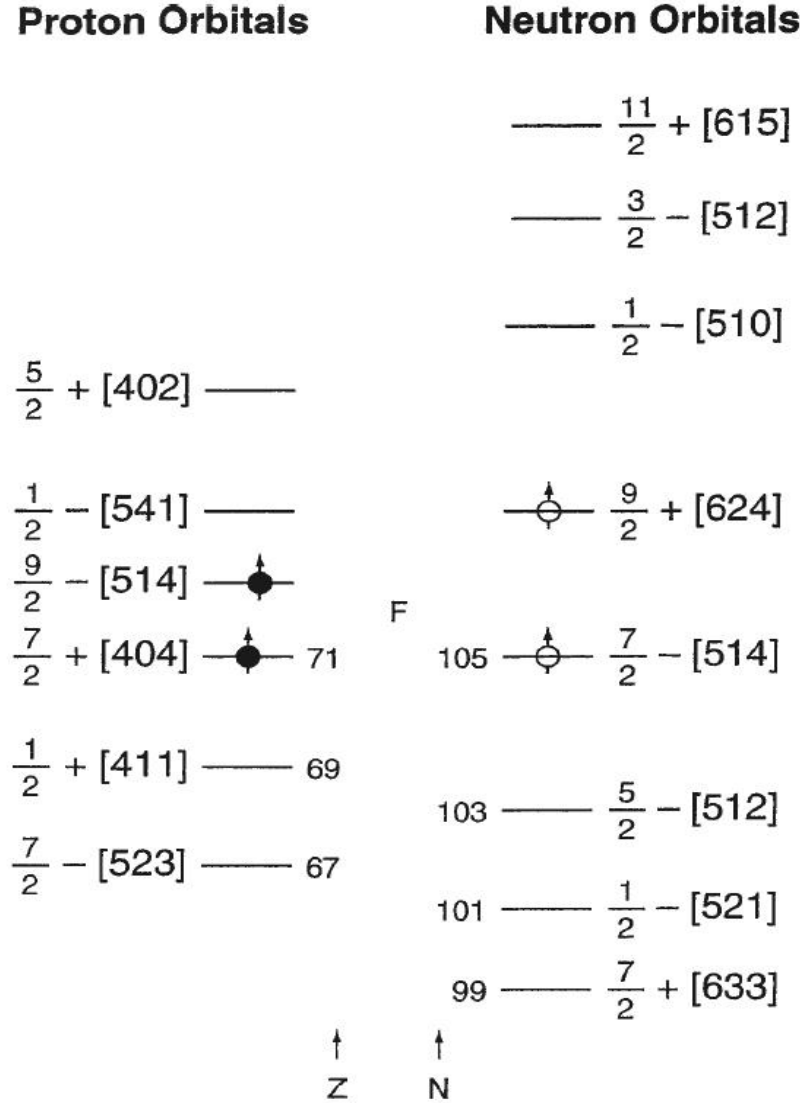


Figure 1. Relative positions of the single-particle orbitals in region of $Z=72$ and $N=106$. The Fermi surface for the neutron and proton system in ^{178}Hf is indicated by the symbol F.

From a phenomenological argument we can have the following cases for a faster decay of the 16^+ state by exciting the isomer to a different level:

If the incoming neutron can excite the proton in a $9/2[514]$ orbital to $5/2[402]$, it will lead to excitation of states of the nearby band with $K^\pi = 14^-$ with the band head at level with energy 2574 keV and half-life of 68 μ s. (see the following, figure 2)

E(keV)	K^π	Single Particle orbital
2446	16^+	$\{7/2[404]_p + 9/2[514]_p + 7/2[514]_n + 9/2[624]_n\}_{16^+}$
2574	14^-	$\{7/2[404]_p + 5/2[402]_p + 7/2[514]_n + 9/2[624]_n\}_{14^-}$

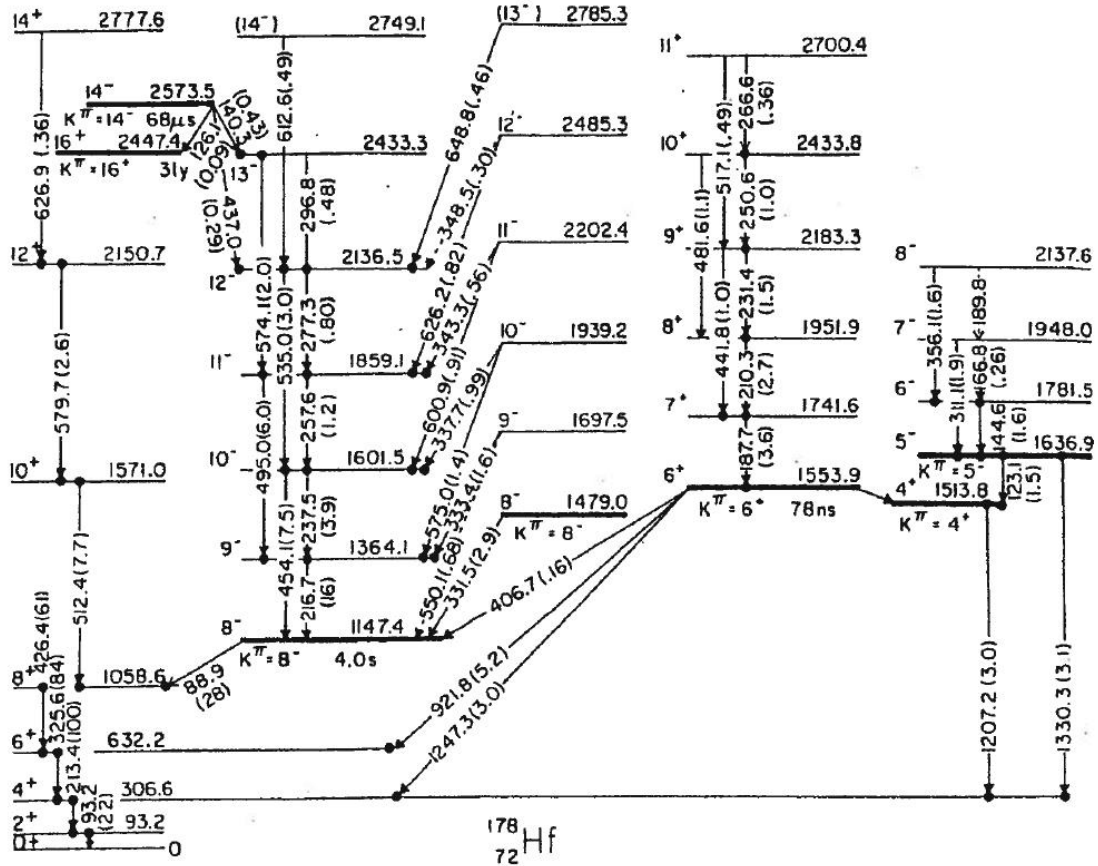


Figure 2. Partial decay scheme from $^{176}\text{Yb}(\alpha, 2n)$ reaction at 26 MeV α energy (T. L. Khoo and G. Lovhoiden, Phys. Lett. 67B, 271, 1977)

A four-quasi-particle state with $K^\pi = 12^+$ based on the configuration
 $\{7/2[404]_p + 1/2(541)_p + 7/2[514]_n + 9/2[624]_n\}_{12}$
or
 $\{7/2[404]_p + 5/2[402]_p + 7/2[514]_n + 5/2[512]_n\}_{12}$

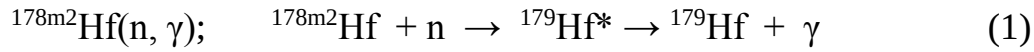
may exist in the neighborhood of the 16^+ and 14^- levels. If the incoming neutron can excite the proton from $9/2[514]$ to $1/2[541]$, or change proton from $9/2[514]$ to $5/2[402]$ and in the same time make a neutron hole in $5/2[512]_n$.

Any one of these three cases can destroy the isomer. We are sure that both 14^- and 12^+ bands have shorter half-lives, and a fast gamma decay will follow. In 2010 T. L. Yang et al⁵ published their calculations of all possible multi quasi-particle states excitation energies, their results showed the possible 12^+ band; it supported our assumption.

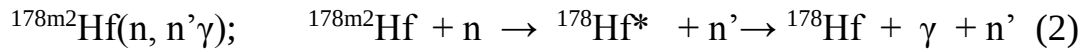
We point out that an incoming neutron can change the nuclear state, but one neutron interaction can only change one isomer nucleus there is almost no energy gain. What we wish is to find a ‘triggering process’ in a chain reaction.

In order to have a fast release of total isomer energy, we need a *sustaining chain-reaction*.

The most interesting case is that the incoming neutron can cause either one of the following two reactions, which will destroy the isomer;



and



When we prepared that article, we neglected to include a 1997 published paper⁶, which reported that for reactions $^{178m2}\text{Hf}(n, \gamma)^{179m2}\text{Hf}$, the measured cross section for thermal neutron $\sigma_{th} = 46 \pm 5$ barns. The $(n, n'\gamma)$ reaction was also mentioned, but no cross section was reported; this provides a third possibility in addition to the two listed above,



The results of reactions (2) and (3) are same, but they go through different intermediate states. However all three reactions represent the destruction of the isomer. In a 2006 publication, Karamian et al⁷ reported,

with natural Hf metal samples irradiated in a reactor, $^{178\text{m}2}\text{Hf}$ could be produced with the following reaction cross sections;

$$\begin{array}{ll} ^{177}\text{Hf}(\text{n}, \gamma) & 2.6 \mu\text{b} \\ ^{178}\text{Hf}(\text{n}, \text{n}'\gamma) & \leq 7 \mu\text{b} \end{array}$$

In the article, they mentioned also “... there are significant cross sections for burn up of $^{178\text{m}2}\text{Hf}$ in the $^{178\text{m}2}\text{Hf}(\text{n}, \gamma) ^{179\text{m}2,\text{g}}\text{Hf}$ reaction equal in total $\sigma_{\text{th}} = 235 \text{ b} \dots$ ”. It is important to point out that the cross section they reported is the difference of production and burn up when the sample was exposed to a neutron beam for a long time allowing the destroy process to occur. The production cross section should also be in order of barns.

All authors in references 6 and 7, did not realize that the destruction of $^{178\text{m}2}\text{Hf}$ isomer is an *effective triggering process*.

One more thing to be emphasized here, is whether the re-emitted neutrons in the $^{178\text{m}2}\text{Hf}(\text{n}, \text{n}'\gamma)$ reaction can induce another $^{178\text{m}2}\text{Hf}(\text{n}, \text{n}'\gamma)$ reaction, if so, there will be “*an effective neutron- induced chain reaction*”.

We can also consider the $^{178\text{m}2}\text{Hf}(\text{n}, 2\text{n})$ reaction. The cross sections for this reaction on all stable isotopes are at barns order with threshold neutron energy of about 8 MeV. We have not seen the reaction on isomer state yet. If it has also a large cross section, and produce neutrons with energy large than 8 MeV, one more neutron produced in this reaction may help for the occurrence of chain-reaction.

On the other hand, gamma-ray induced fast de-excitation was one of the controversies, mainly because the experiment detected a very low intensity gamma transition associated with $^{178\text{m}2}\text{Hf}$ decay, which implies a very small interaction cross section, or with wrong energy of the incident gamma rays. However no matter how small the interaction cross section is, as long as gamma ray can induce the transition by whatever the mechanism, the de-excitation gamma rays with a broad Compton spectrum would induce further de-excitation, and a *self-sustaining chain-reaction* would occur when the amount of $^{178\text{m}2}\text{Hf}$ nuclei reaches a ‘*critical mass*’.

PROPOSED EXPERIMENTS

In all $^{178m2}\text{Hf}$ production methods; such as $^{177}\text{Hf}(n, \gamma)$ in reactor irradiation, $^{179}\text{Hf}(n, 2n)$, and $^{176}\text{Yb}(\alpha, 2n)$ reaction with accelerators, as long as involving neutrons in the reaction, either as incident particles, or as by-product, either one of the following reactions $^{178m2}\text{Hf}(n, \gamma)$, $^{178m2}\text{Hf}(n, n'\gamma)$ process will destruct the isomers. The reported production cross sections in previous articles actually were the difference of the production and destruction. To avoid the destruction process, we should not use a long time reactor irradiation, or an accelerator beam for reaction process.

We propose to use a very intense but short neutron pulse to irradiate our target. In principle, the first time we send the neutron pulse to target, there is the creation process, with a very small chance of destruction. By measuring the gamma spectrum of isomer decay, we can determine the number of isomer nuclei produced, and calculate the production cross section at that neutron energy. The second time we send the neutron pulse to the same target, the activity should be twice, but if the destruction process occurs, the activity will be less than twice. If we repeat the experiments, the activity curve will deviate from a straight line. From the deviation of the straight line, we can deduce the destruction cross section at that neutron energy.

In last few years, Los Alamos National Lab has had a collaborative program with Livermore National Lab, ARE of United Kingdom, Plasmas Science and Fusion center of MIT, and the University of Rochester. The purpose is to study the characteristics of the DT fusion reaction occurring in a laser-induced implosion of a DT capsule. Many results were published. One of us (HH) is part of the team studying gamma rays produced in the DT reaction^{8,9,10}. The 14.1 MeV neutrons produced in this reaction satisfy our need for a “yes or no” experiment we propose here. The neutron pulse, produced at the OMEGA laser system (University of Rochester), has a pulse width of 100 – 200 ps, and total neutron number of $\sim 10^{14}$. Both parameters depend on the target capsule, its DT densities, and burn temperature. If we use the same kind of capsule in all our measurement, the uncertainty of reproducibility is within 10%.

We can add a Compton detector to the set-up, to measure the amount of gamma rays produced in the process of 14-MeV neutrons interacting with the target, mainly the (n, γ) reaction of Hf isotopes, and gamma rays

produced in the capsule and surrounding materials. If the incoming neutron can destruct the isomer, in the second time when neutrons bombard the target, the Compton detector will detect more gamma rays, which are from the isomer decay. This is evidence of neutron induced destruction process.

If we can produce the isomer successfully, and accumulate more isomer sources, we will perform the following measurements to establish whether gamma ray can induce destruction process.

Because we do not know what energy of gamma ray we should use, we propose to use electron produced Bremsstrahlung (up to about 500 keV energy, this is the energy range of decay gamma rays) as our photon source;

1. Expose the isomer sample to the Bremsstrahlung beam, if the activity of the sample after the exposure is smaller than the activity before the exposure, there is net destruction.
2. If the Compton detector detects more gamma rays, with an isomer target exposed to Bremsstrahlung than the case with a blank Hf target, this will be the evidence of gamma-ray induced destruction process.

The gamma rays produced in isomer decay may, in turn, cause chain reactions

CONCLUSIONS

We postulate the following conclusions that the success of the proposed experiment will provide:

1. An effective method to produce $^{178m2}\text{Hf}$ isomers. Up to now, most isomers were produced in reactor, or chemical separation from an accelerator beam stop. If our argument is correct, the destruction process also occurs. A strong and short pulse of neutrons is a better way to produce the isomer.
2. To establish the existence of the destruction process, the $(n, n'\gamma)$ reaction reported in reference 7 is the only report so far, that shows that neutron can 'destroy' the isomer. If we can successively carry out our experiment, it will verify the destruction process.

3. The possibility of an isomer nuclei chain reaction. If we want to release all isomer energy, a chain reaction is needed. We expect that the cross section for neutron induced chain reaction would be larger than that by gamma rays, but the later one is more interesting, because it may establish a '*critical mass*', no matter how large it will be.
4. There exist many nuclear isomers with reasonable life time, we can try the experiment method to other isomers in both creation and destruction.

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