Applications; Role of nuclear physics research in nuclear nonproliferation, stewardship and homeland security



Ed Hartouni

Exotic Beam Summer School 2011 NSCL, MSU, East Lansing MI August 19, 2011

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Security, LLC, Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Background and outline

Ed Hartouni:

Trained in experimental High Energy Physics at Columbia University, collaborated on experiments at SLAC, Fermilab, BNL, CERN Taught Physics at UMass Amherst from 1986 to 1995 Moved to LLNL in 1995, was leader of N-Division from 2001 to 2008 (Alter-ego active in the sport of climbing natural vertical geographic features)

Outline

- Overview
- Introduction to the examples

four examples:

- Stockpile Stewardship Neutron reactions on unstable nuclei
- Homeland Security Signatures of Highly Enriched Uranium (HEU)
- Policy & Advice Hf isomers and their potential uses
- Energy National Ignition Facility (NIF) and inertial confinement fusion (ICF)



Overview

OFFICE OF SCIENTIFIC RESEARCH AND DEVELOPMENT 1530 P Street, NW. Washington 25, D.C. JULY 25, 1945

DEAR MR. PRESIDENT:

In a letter dated November 17, 1944, President Roosevelt requested my recommendations on the following points:

(1) What can be done, consistent with military security, and with the prior approval of the military authorities, to make known to the world as soon as possible the contributions which have been made during our war effort to scientific knowledge?

(2) With particular reference to the war of science against disease, what can be done now to organize a program for continuing in the future the work which has been done in medicine and related sciences?

(3) What can the Government do now and in the future to aid research activities by public and private organizations?

(4) Can an effective program be proposed for discovering and developing scientific talent in American youth so that the continuing future of scientific research in this country may be assured on a level comparable to what has been done during the war?

It is clear from President Roosevelt's letter that in speaking of science that he had in mind the natural sciences, including biology and medicine, and I have so interpreted his questions. Progress in other fields, such as the social sciences and the humanities, is likewise important; but the program for science presented in my report warrants immediate attention.

In seeking answers to President Roosevelt's questions I have had the assistance of distinguished committees specially qualified to advise in respect to these subjects. The committees have given these matters the serious attention they deserve; indeed, they have regarded this as an opportunity to participate in shaping the policy of the country with reference to scientific research. They have had many meetings and have submitted formal reports. I have been in close touch with the work of the committees and with their members throughout. I have examined all of the data they assembled and the suggestions they submitted on the points raised in President Roosevelt's letter.

Although the report which I submit herewith is my own, the facts, conclusions, and recommendations are based on the findings of the committees which have studied these questions. Since my report is necessarily brief, I am including as appendices the full reports of the committees.

A single mechanism for implementing the record committees is essential. In proposing such a somewhat from the specific recommendations of have since been assured that the plan I am pracceptable to the committee members. 66 years ago Vannevar Bush provides recommendation of regarding the science enterprise in the post-war United States.

The resulting report "Science – The Endless Frontier" laid out the structure of US science funding, created the NSF and eventually led to the National Laboratory system.

The importance of the science and of the connection of that science to the "application" was recognized in Bush's recommendations to the president.

The pioneer spirit is still vigorous within this nation. Science offers a largely unexplored hinterland for the pioneer who has the tools for his task. The rewards of such exploration both for the Nation and the individual are great. Scientific progress is one essential key to our security as a nation, to our better health, to more jobs, to a higher standard of living, and to our cultural progress.

Respectfully yours,

(s) V. Bush, Director

THE PRESIDENT OF THE UNITED STATES, The White House, Washington, D. C.



Introduction – policy

Policy, from Wikipedia:

"A policy is typically described as a principle or rule to guide decisions and achieve rational outcome(s)."

Generalized statement:

"A policy is created through the process of making important organizational decisions, including the identification of priorities and different alternatives such as personnel and technology programs or budget priorities, and choosing among them on the basis of the impact they will have on the overall development. Policies can be understood as political, management, financial, administrative and executable mechanisms arranged to reach explicit organizational goals and objectives."

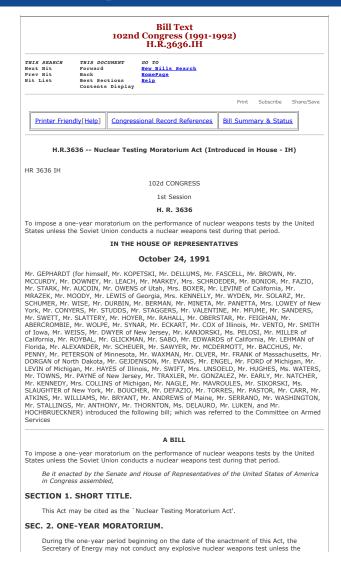
Capability management:

"Capability management aims to balance economy in meeting current operational requirements, with the sustainable use of current capabilities, and the development of future capabilities, to meet the sometimes competing strategic and current operational objectives of an enterprise."

Science and technology are a part of the policy making process



Stockpile Stewardship: Nuclear science in support of the Comprehensive Test Ban Treaty (CTBT)



1992 "Nuclear Testing Moratorium Act"

SEC. 2. ONE-YEAR MORATORIUM.

During the one-year period beginning on the date of the enactment of this Act, the Secretary of Energy may not conduct any explosive nuclear weapons test unless the President certifies to Congress that the Soviet Union (or a successor state of any part of the Soviet Union) has conducted an explosive nuclear weapons test during that period.

1996 the US signed the CTBT but did not ratify.

The US has remained in compliance with the CTBT.

We have implemented a stockpile stewardship program to replace the need for testing.

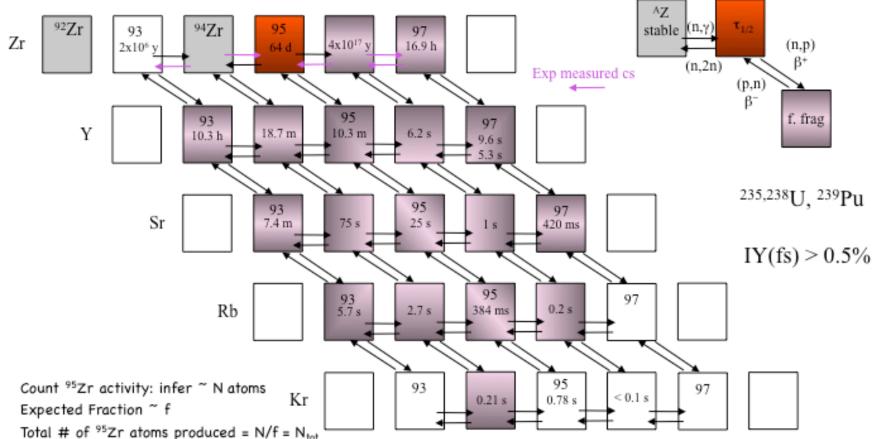
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Neutrons, tracers and radiochemistry



- Total # of fissions F = Ntot /0.06 ~ M fissions
- NOTE: 0.06 is the percentage of the total fragment distribution that is assumed to decay along the A=95 mass chain into 95Zr. IT ASSUMES NO BURN IN OR OUT OF THE CHAIN.
- Energy release from fission ~ FY x 1023 fissions/kt

TOTAL FISSION YIELD = F / Ax1023 kt

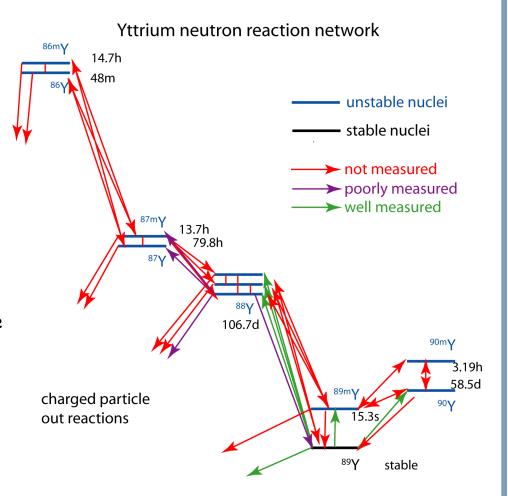
Reactions on unstable nuclei

A known quantity of a specific isotope (e.g. ⁸⁹Y) is prepared and irradiated by an unknown flux of neutrons.

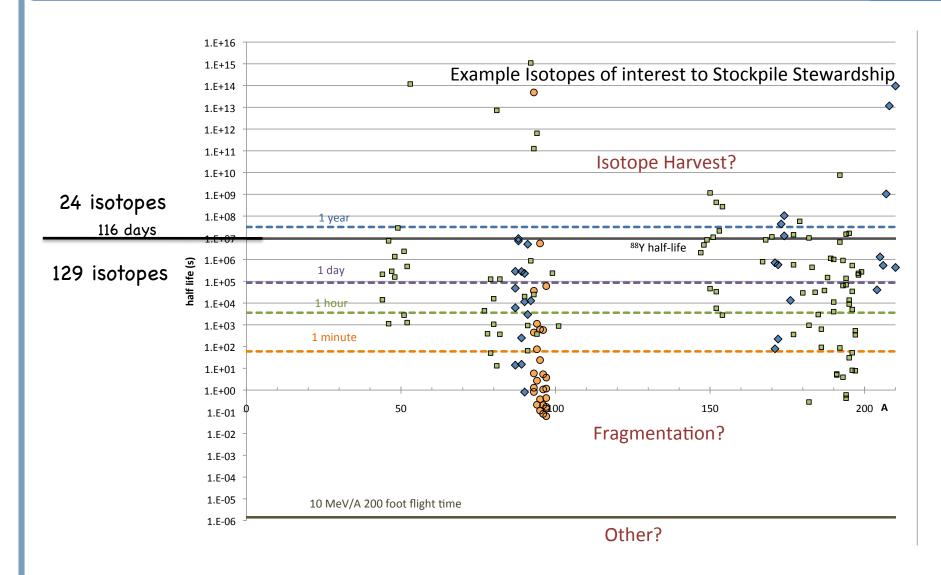
Samples of the element are recovered and the ratio of isotopes (e.g. ⁸⁸Y/⁸⁹Y) are used to infer information regarding the neutron flux.

Understanding the observed ratio requires physics input, such as the rate of producing and destroying the isotopes appearing in the ratio (including isomers), all as a function of neutron energy.

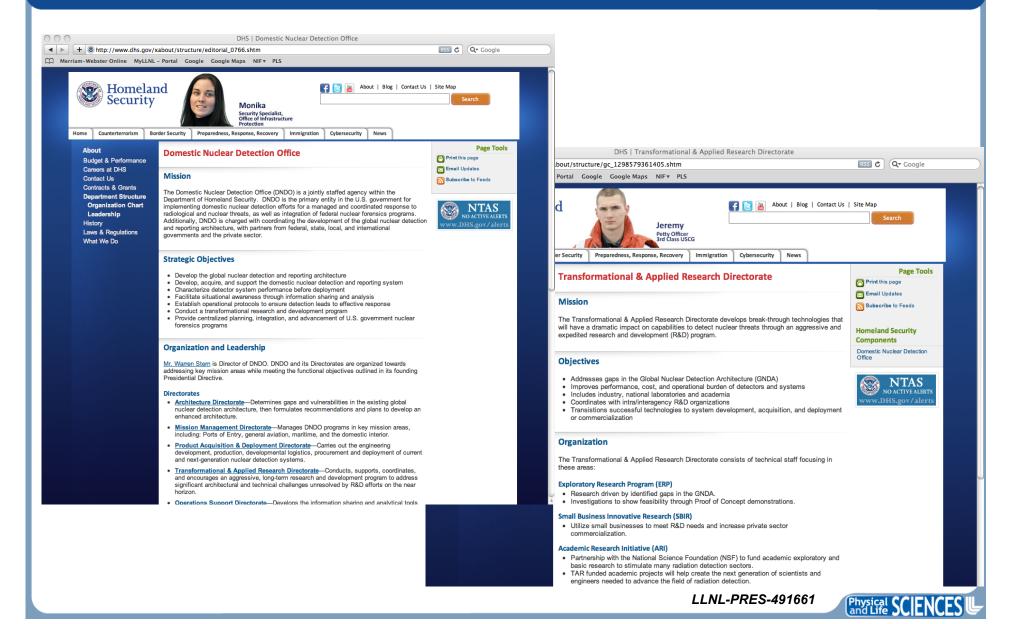
In the case of Yttrium, of the 64 relevant cross sections, only 7 are measured, the rest are estimated.



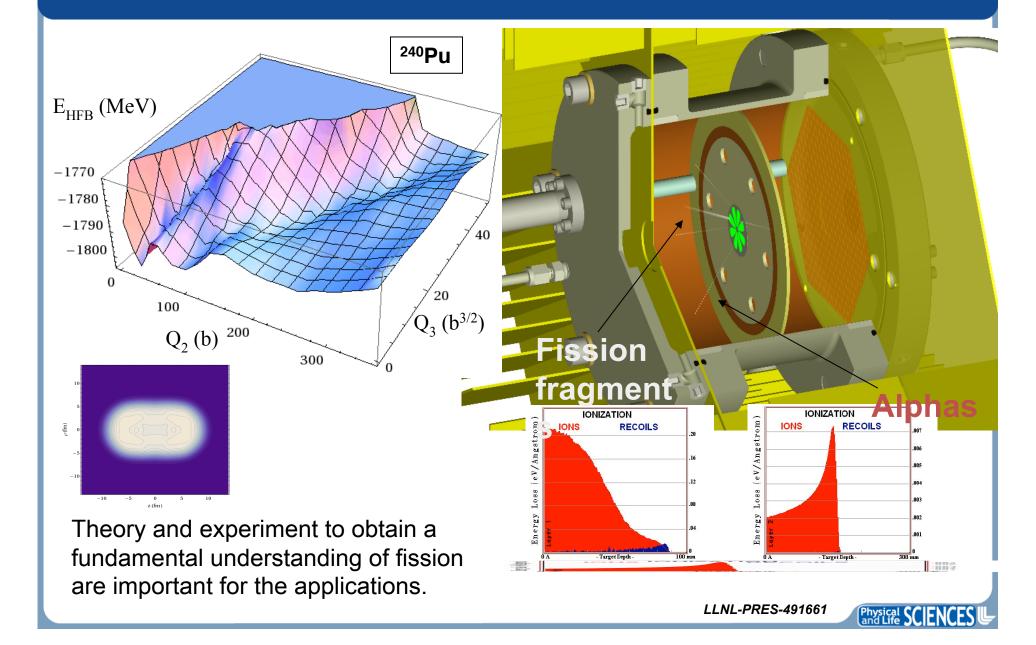
Filling the gaps in nuclear evaluations



Homeland Security: Signatures of Highly Enriched Uranium



Fission

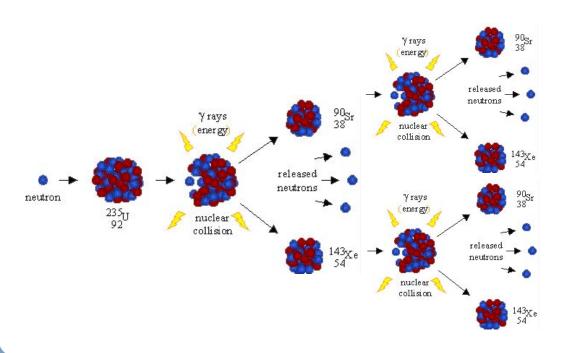


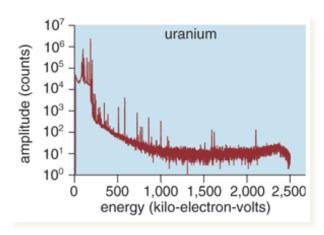
Chain-reactions symptoms and signs

Radioactive decay is an *independent* process – Poisson distribution

Radiation resulting from fission chains is correlated – not Poisson

Special Nuclear Material (SNM) has as its most important attribute (in this application) the ability to sustain fission chains. It is also radioactive but not necessarily a characteristic that discriminates from other radioactive material.







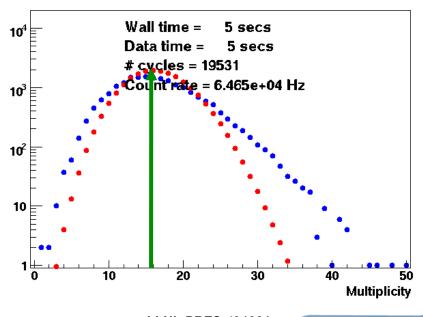
Feynman variance

Count neutron pairs within a time bin subtract random (Poisson) expectation divide by mean call this <u>Feynman Variance</u>

Form similar 3,4-neutron coincident (moments or cumulants)

[<n(n-1)>
- <n²>]
÷<n>
= R2F
R3F,R4F

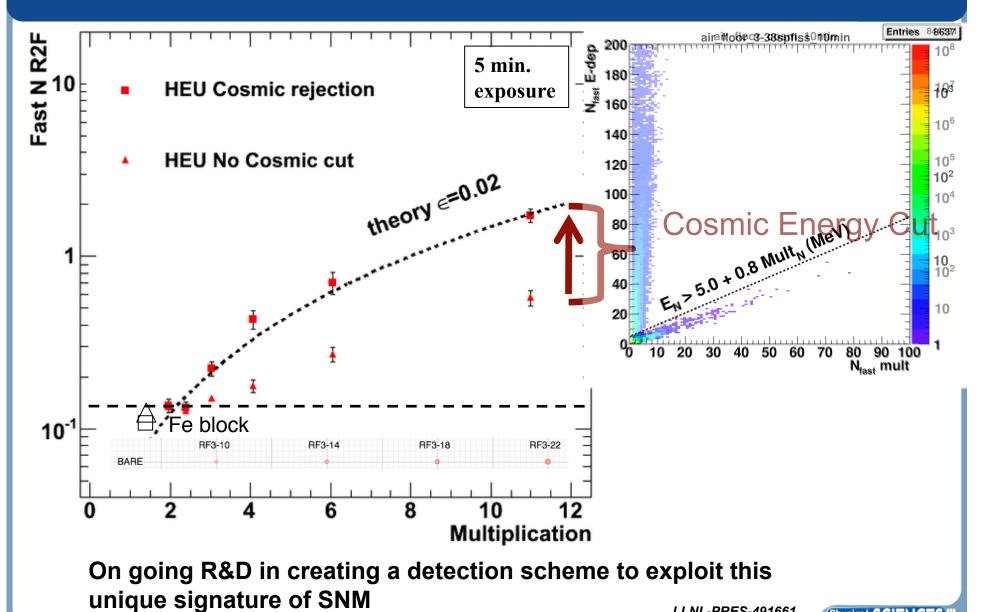
Backgrounds would have to "fake" a time correlation with the neutrons, e.g. cosmic ray interactions in the surrounding materials



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Possible application



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Policy and Advice: "What about Hf isomers?"

A request from DOE to help it respond to questions from Congress

Draft Terms of Reference for a literature review and assessment of the state of research on Nuclear Isomer ^{178m2}Hf and the potential for controlled energy release.

Background: Nuclear isomers have long been proposed as novel High Energy Density Materials (HEDM). The former Soviet, Russian and US literature all contain published articles on the production of nuclear isomers and discussions on potential stimulated deexcitation mechanisms. Further, US research programs since approximately 2001 have unsuccessfully attempted to validate the HEDM nature and controlled energy release from nuclear isomers. Recently, the Senate Armed Service Committee (SASC) staff tasked the NNSA Office of Nonproliferation Research and Development (NA-22) to conduct an all-source literature review and produce a short written report that provides the current scientific assessment of the potential for controlled energy release from the nuclear isomer ^{178m2}Hf for energy and military uses, but the mechanism of enhanced decay through neutron inelastic scattering interactions. SASC staff request that the report be completed by the end of September 2008.

Key Research Question: Does either current scientific theory or previous experimental results support the claims made by the recent Russian literature that neutron induced stimulated de-excitation through inelastic scatter produces enhanced decay of the nuclear isomer ^{178m2}Hf, potentially providing substantial and controllable energy release?

Product: Written, all-source report, including citation index, that answers the research question above and provides supporting information.



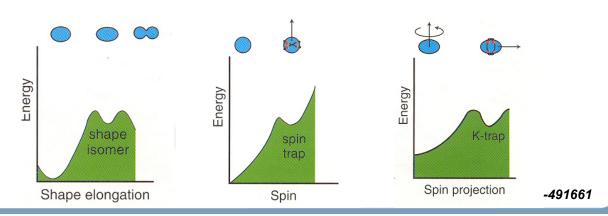
What's a nuclear isomer?

Frederick Soddy anticipated nuclear isomers in a 1917 discourse delivered to the Royal Society:

nuclei with the same atomic number and atomic charge but with distinct radioactive decay properties.

A more modern view consistent with Soddy would define nuclear isomers as states of a nuclei with anomalously long life-times, due to forbidden or "hindered" decays to the ground state, from ~10 ps to 10³¹ y.

Nuclei are n-body quantum systems which exhibit both single-particle and collective behavior. Isomers exist as: "shape" isomers, spin isomers, *K*-isomer.



How was ^{178m2}Hf discovered?

From: "Charles Reich" <cwreich@xxx.yyy>
To: "Edward P. Hartouni" <hartouni1@llnl.gov>
Subject: Re: history of 178m2Hf discovery
Date: Sat, 31 May 2008 15:45:30 -0600

Dear Dr. Hartouni,

Thank you very much for your recent query and the interest it demonstrated in the 31-y isomer in 178Hf. The short answer to your question regarding our discovery of this unique state is simply "serendipity". I would like to claim some superior insight or wisdom, but unfortunately I cannot. As I remember it from this vantage point of approximately 40 years after the event, it went something like this. We were located in the vicinity of two of the highest-flux reactors in the country (world?) and decided to insert some samples into them, simply leave them in for an extended time, and "just see what happened". With Hf, we struck "pay dirt". We were initially puzzled by the data, but in due course were able to come up with a unique, and convincing, decay scheme for the activity. At that point, it was clear that we had something truly exciting; and I spent considerable time and effort trying to extract from our data as much information that I felt was reasonably implied by them.

Please note that we considered ourselves to have been "lucky". If, for example, the half-life had been significantly shorter, we would probably not have been able to observe the isomer. However, it wasn't, and we did. Also, we naively assumed that the isomer had been formed by a neutron-capture reaction. Subsequently, we came to believe that it was more likely produced by an (n,2n) reaction (although this may still be an open question).

I trust that this has been responsive to your request. If not, please let me know and I will try to answer any other questions that you may have.

Now, questions for you! I was not aware of your project and would like to know more about it. Does it mean that there is still interest in the "Hf bomb"effort, or does it have a broader focus?

As your project proceeds, I would appreciate being kept up to date on it.

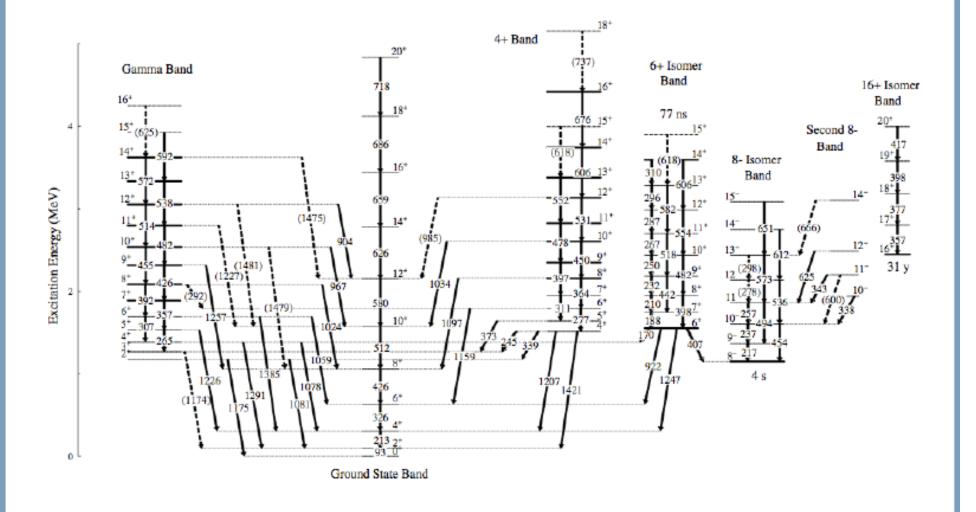
Best wishes, and thank you for your interest.

Charles Reich

100 mg of HfO₂ irradiated for 2 years and allowed to decay for 3 years yielded ~25 pg of ^{178m2}Hf



¹⁷⁸Hf level scheme



Isomers, Gamma-ray lasers, controversy

Collins, et al. Physical Review Letters 82, 495 (1999)

A sample of 6.3×10^{14} nuclei of the 4-quasiparticle isomer of 178 Hf having a half-life of 31 yr and excitation energy of 2.446 MeV was irradiated with x-ray pulses from a device typically used in dental medicine. It was operated at 15 mA to produce bremsstrahlung radiation with an end point energy set to be 70 or 90 keV. Spectra of the isomeric target were taken with a high purity Ge detector. Intensities of selected transitions in the normal decay cascade of the ¹⁷⁸Hf isomer were found to increase by about 4%. Such an accelerated decay is consistent with an integrated cross section of 1×10^{-21} cm² keV for the resonant absorption of x rays to induce gamma decay. [S0031-9007(98)08333-1]

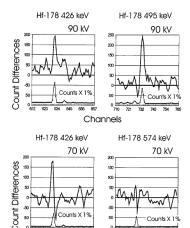


FIG. 2. Plot of the differences in counts obtained in the spectrum of the ¹⁷⁸Hf from the isomeric target when it was exposed to the x-ray beam and when it was not. A weighted smoothing procedure was applied. Also shown are the unsmoothed spectra from the unirradiated target scaled by 1% to facilitate comparison. In the upper row from left to right, the two lines correspond to the transitions $(8^+ \rightarrow 6^+)$ and $(11^- \rightarrow 9^-)$, shown by the heavy arrows in Fig. 3. The lower row from left to right presents again the $(8^+ \rightarrow 6^+)$ transition together with the $(13^- \rightarrow 11^-)$.

Channels

Spontaneous decay of target isotopes must be subtracted, beam on beam off.

Decay is "delayed" by the 4 s isomer.

Reported cross section is 6 orders of magnitude of levels relevant to this experiment. The spontaneous decay of the 31-yr, 16^+ isomer is shown by the thin arrows. The higher than expectation.

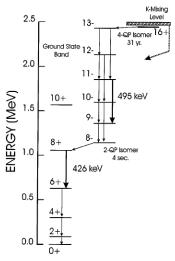


FIG. 3. Energy level diagram for ¹⁷⁸Hf showing a selection thick solid arrows show components studied in this work from the particular cascade from the decay forced by the x-ray irradiation. Heavy dotted arrows are inferred transitions needed to feed those shown.

from Collins et al. Physics of Atomic Nuclei 63, 2067 (2000)

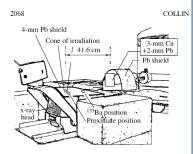


Fig. 1. Scheme of the experimental arrangement showing the geometric placements and dimensions of the compo

Further research is needed to provide greater precision to the measurements of the transition energy to the K-mixing level and to clarify properties of the cascade feeding the GSB. Such data will then facilitate a better understanding of these first evidences of the triggering of induced gamma emission from the 31-yr isomer of ¹⁷⁸Hf with very low energy photons through large integrated cross sections, $\sigma\Gamma \ge 2 \times 10^{-23}$ cm² keV.



Science follow-up

Ahmad, et al., Physical Review Letters, 87 072503 (2001)

Enhanced decay of the 31-yr isomer of 178 Hf induced by x-ray irradiation has been reported previously. Here we describe an attempt to reproduce this result with an intense "white" x-ray beam from the Advanced Photon Source. No induced decay was observed. The upper limits for the energy-integrated cross sections for such a process, over the range of energies of 20-60 keV x rays, are less than 2×10^{-27} cm² keV, below the previously reported values by more than 5 orders of magnitude; at 8 keV the limit is 5×10^{-26} cm² keV.

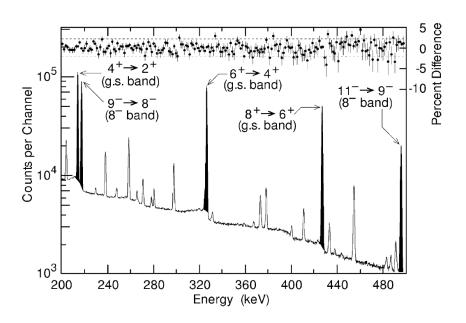


FIG. 1. Energy level diagram showing the decay of the 31-yr ¹⁷⁸Hf isomer. The transition energies are labeled in keV. Those transitions that were reported to be enhanced in [3,6] are highlighted.

no "triggering" observed, consistent with expectations from theory.

No independent experimental confirmation.

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"...claims made in recent Russian literature..."

RUSSIAN SCIENTIFIC CENTER

WKURCHATOVINSTITUTE"

G.V. MURADIAN

443-6310

DECAY OF 178m2Hf ISOMER NUCLEI DURING NEUTRON

INELASTIC SCATTERIN



Moscow — 2004

УДК 539.143

Key words: isomer, hafnium, neutron, inelastic scattering, decay of isomer nuclei.

It is shown that the neutron can initiate the chain self-sustaining process of ^{178m2}Hf isomeric nuclei energy release. The experiments on search for other isorners which are more suitable for the chain process are proposed.

Isomer 178^{m2}Hf represents a high-spin (J=16) excited state of the nucle is ¹⁷⁸Hf. It is characterized by high energy of excitation of 2.45 MeV and by a large decay period of 31 years. During the decay the energy of the isomer converts mostly into γ-radiation. Along with spontaneous decay the isomer could also break down instantly by the action of charged particles, γ-quantum and neutrons.

Decay by the action of neutrons may be the result of inelastic scattering. After such scattering the nucleus appears in a different (non-isomeric) state and breaks down (in case this is not the ground state) by way of passing into lower states. In the nedium that consists of someric nuclei, a neutron that is emitted as a result of the first inelastic scattering, could, in the similar manner, cause a decay of the next isomeric nucleus and so forth. In this way, a neutron initiates consecutive decay of isomeric nuclei.

Nevertheless, sooner or later, this process will come to an end as the result of radiation capture or leak of scattered neutrons. To provide continuity of the decay process, the iso neric nuclei could be placed into the medium that consists of the Beryllium nuclei. Because of the reaction ⁹Be(n,2n) the loss of neutrons is compensated; so the chain of isomeric nuclei decay is not broken. The threshold of the reaction ⁹Be(n,2n) is 1.84 MeV. The energy of the neutron could exceed this threshold since the neutron could a cquire additional energy during scattering on the isomer unlike during scattering on regular nuclei super elastic scattering).

This is a new type of nuclear chain reaction. It is significantly different from the only known nuclear chain reaction that is based on a nuclear fission. In the case of isomers, there is no formation of radioactive nuclei and the major portion of energy is released as γ-rediation.

For the chain reaction on the isomer to proceed, it is necessary that the cross section of inelastic scattering was large enough as compared to the cross section of neutron absorption and also that the spectrum of inelastic scattered neutrons contained a sufficiently large fraction of neutrons that are beyond the threshold of the reaction.

9Be(n,2n).

data for these cross sections do not exist. Their evaluation because the isomer, in its structure, sharply differs from an

difficult exp ed in Kurch mportant fo the work [een the reso capture and e that radia of existing s e is also sm ique in the tion ⁹Be(n,2 his manner, sorption, and d, the condit e seen from

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te experime cross section states that for ghlight of a scludes a poisomer staig would pri

- 3. The weight of the states with the assigned quantum number K in compound-states grows with the decrease of K. Since the states of ¹⁷⁸Hf are characterized by relatively low values of K, it could be expected that the share of inelastic-scattered neutrons will increase with acquisition of energy.
- 4. The isomer resonances are characterized by large full cross sections. Thus, the resonance of 0.75 eV has a cross section of $\sim 10^5$ barn. The value of resonance for thermal neutrons is also large. The large cross section could foster a decrease in the quartity of isomer nuclei that is required for the chain reaction to proceed.

Notice that other nuclei could be introduced along with the Beryllium in order to optimize the chain reaction. For example, a small addition of fissile nuclei could substantially improve the balance of neutrons with insignificant amount of radionuclides that are formed during fission.

The consideration of a possibility of a chain reaction that uses isomer nuclei decay was made above on the example of 178^{m2}Hf, which is the most suitable among the known isomers. However, it is very likely that there exist some other more suitable isomers that could be produced relatively easily by bombardment in the reactor or the accelerator, found among fission fragments or even found in Nature. It can't be ruled out that those will be isomers of shape, isomers of density (super-dense nuclei) and in general, isomers of unknown until now nature.

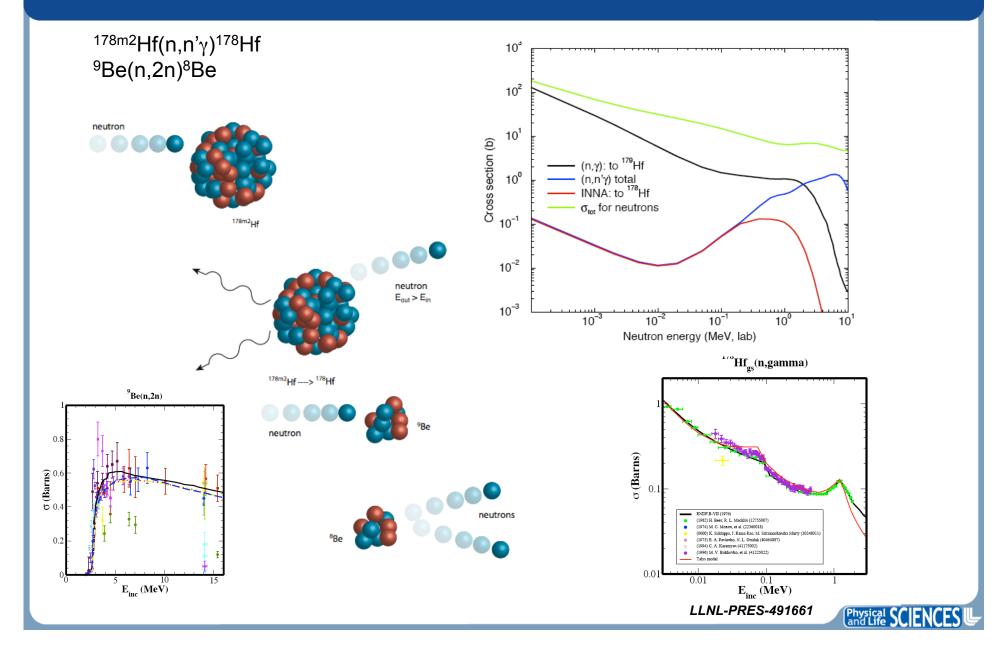
The search for suitable isomers regardless of their nature could be conducted directly during experiments that observe accelerated neutrons undergoing inelastic scattering. The spectrum of these neutrons provides a direct answer to the question about a possibility to conduct a chain reaction of decay of isomer nuclei. The new isomers could be detected and studied also by a way of identifying and researching neutron resonances of appropriate targets.

The Author expresses his appreciation to Academician S. T. Belyaev for his fruitful discussions and support.

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A new type of chain reaction?



Evaluation of the likelihood of isomer applications

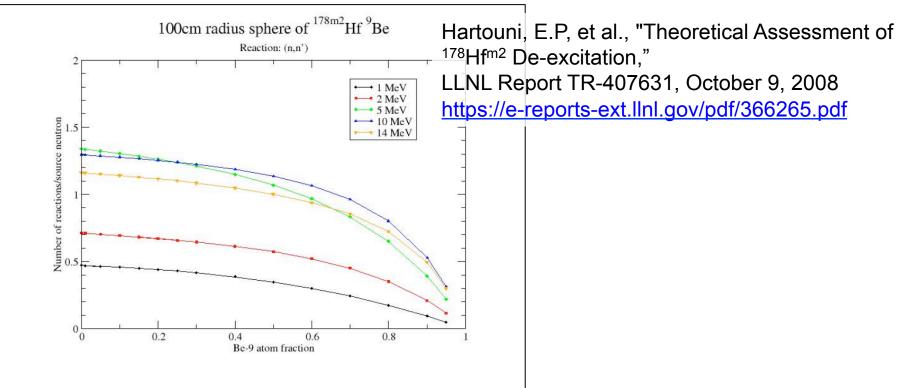


Figure 1. Effect of Beryllium concentration on the simulated (n,n') reactions per source neutron in a 100cm sphere of 178m2 Hf and 9 Be with a point source located at the center of the sphere. Results are shown for five neutron energies and can be found in Table I.

"Our conclusion is that the utilization of nuclear isomers for energy storage is impractical from the points of view of nuclear structure, nuclear reactions, and of prospects for controlled energy release. We note that the cost of producing the nuclear isomer is likely to be extraordinarily high, and that the technologies that would be required to perform the task are beyond anything done before and are difficult to cost at this time."



Lessons learned

"I say that's also important in giving certain types of government advice. Supposing a senator asked you for advice about whether drilling a hole should be done in his state; and you decide it would be better in some other state. If you don't publish such a result, it seems to me you're not giving scientific advice. You're being used. If your answer happens to come out in the direction the government or the politicians like, they can use it as an argument in their favor; if it comes out the other way, they don't publish it at all. That's not giving scientific advice."

Richard Feynman 1974 Caltech commencement address http://www.lhup.edu/~DSIMANEK/cargocul.htm

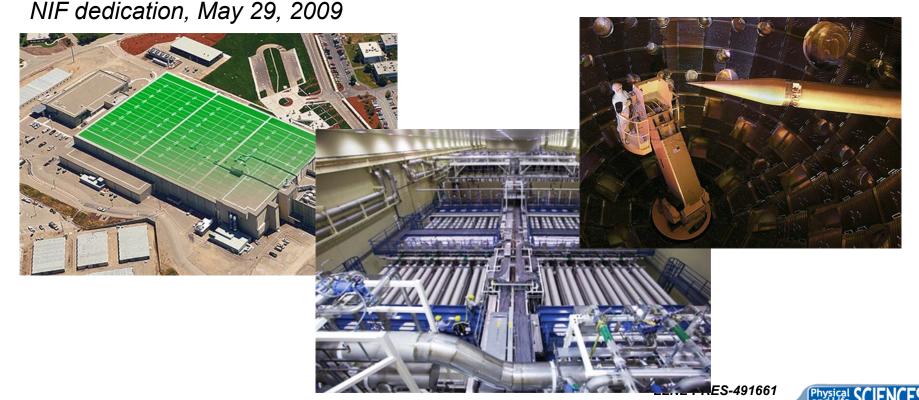


Energy: The National Ignition Facility (NIF)

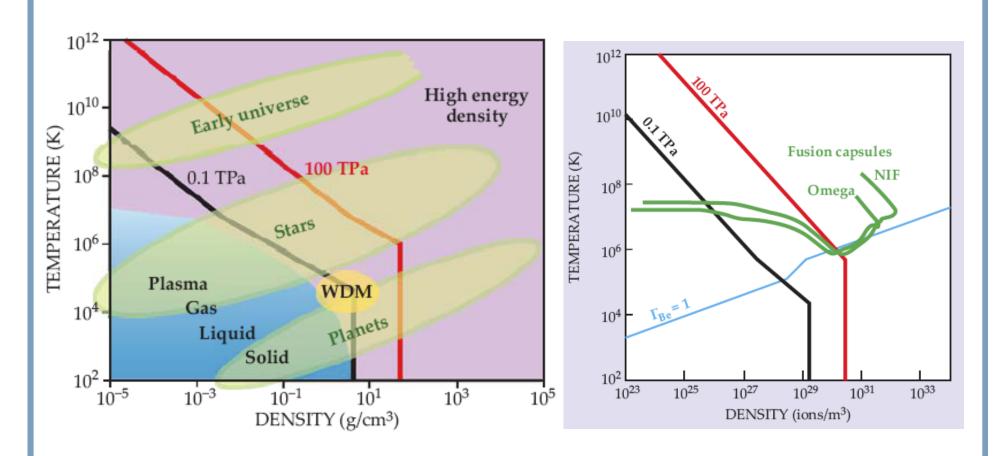
"A successful demonstration of ignition and energy gain at NIF would be a transforming event that would solidify fusion's potential as an important energy source. The dedication of NIF is a milestone in an exciting scientific journey that will create a lasting legacy of discovery, innovation, and security and allow the nation to reap the benefits of this visionary investment in its future,"

Steven Koonin, Under Secretary for Science, U.S. Department of Energy

NIF dedication, May 20, 2000



High Energy Density (HED) science



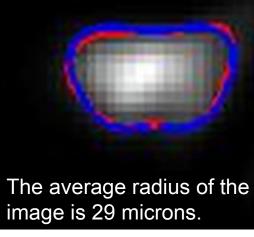
R. Paul Drake, "High-energy-density physics" Physics Today June 10, 2010 page 28

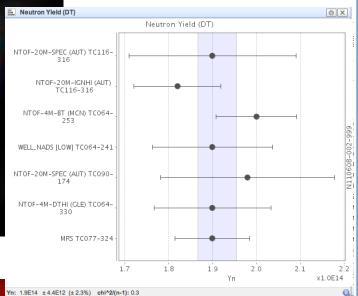


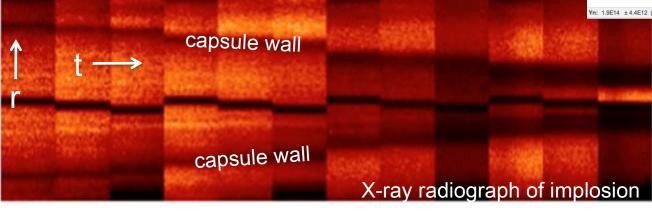
Diagnosing the implosions

A NIF target capsule with the cryogenic DT fuel layer highlighted in violet.

Neutron flux from the first DT shot on June 9 as imaged by the Neutron Imager diagnostic.







Physical SCIENCES

The road forward...

The NIF "ignition campaign" is full of all the grand surprises that any journey into the frontier should offer. The multidisciplinary, international collaboration is trying to do something new and the outcome is important.

"We choose to go to the moon. We choose to go to the moon in this decade and do the other things, not because they are easy, but because they are hard, because that goal will serve to organize and measure the best of our energies and skills, because that challenge is one that we are willing to accept, one we are unwilling to postpone, and one which we intend to win, and the others, too."

John Kennedy – Rice University 1962



Concluding remarks

My very personal approach to this lecture was not to exclude the wonderful, important and interesting work done at many other laboratories, both within the DOE complex and other national and international laboratories. And beyond the realm of national and international security.

Basic nuclear science is an essential foundation from which we build many important applications. Not only the knowledge, but also the people who have that knowledge. Those people are the most important resource.

I wish you all as long and varied and interesting career as I have had. I look forward to seeing your good work.

