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14. ABSTRACT

Nuclear spin isomers store electromagnetic energy at densities reaching 1.3 GJ/g for shelf lives approaching centuries. The best combination of shelf life and energy storage is found in Hf-178. It has been shown to release its stored energy when triggered by x-ray flashes at modest power levels. In this work trigger pulses were derived from tunable, monochromatic x-rays produced by synchrotron radiation sources. It was proven that the triggering was initiated by photoionization of an electron from the L-shell surrounding the isomeric nucleus. A fraction of 0.16% of those photoionizing events led to triggering of the release of the energy stored by the Hf-178 nuclear spin isomer. Confidence in these positive experimental results reached 92 sigma which is equivalent to 1,824 nines of certainty.

15. SUBJECT TERMS

Nuclear spin isomers, High energy densities, Directed energy

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FINAL TECHNICAL REPORT

to the

US AIR FORCE OFFICE OF SCIENTIFIC RESEARCH (AFOSR)

describing accomplishments in the

Renewal of Research on Triggering Nuclear Spin Isomers

during the period
March 01, 2001 to August 31, 2005

supported by

Grant No: F49620-02-1-0141

to the

Center for Quantum Electronics
University of Texas at Dallas
PO Box 830688, Richardson, TX 75083-0688

submitted by the Principal Investigator
Carl B. Collins

November 30, 2005

EXECUTIVE SUMMARY

Nuclear spin isomers store the highest densities of energy possible without nuclear reactions. For example, in the best case one isomer of ^{178}Hf stores 2.445 MeV per atom for a shelf life of 31 years. In practical terms this means that a sample of the size of a golf ball would store the energy equal to 10 tons of chemical fuels or explosives. However, in such nuclear spin isomers the energy is stored electromagnetically so that it would be released as x-rays and γ-rays, if it could be triggered. Prior to our work it had been thought to be impossible to trigger the release of these great energy densities trapped in the internal electromagnetic excitations of nuclear spin isomers.

Since 1988 we had studied the means through which pulses of X-rays could be used to accelerate the radioactive decay of the nuclear spin isomer of ^{180}Ta, a useful vehicle for development, but without practical levels of energy gain. Only 2.7% more energy is released than the amount that is needed for triggering that isomer. Because it was the only nuclear isomer for which samples were available, it was the first to be successfully triggered. Despite the small energy gain, the significance was great because that first success showed that some energy gain was possible.

A decade elapsed before samples of the best of the candidate nuclear isomers became available. In 1998 we succeeded in triggering the first release of the stored energy of the ^{178}Hf isomer [1] and in the subsequent years, January 1, 1999 to November 30, 2001 further progress was supported by an AFOSR Grant to which this report describes a continuation. We showed that the x-rays from a small device familiar from dental examinations proved to be sufficient to trigger the release of the stored energy. In fact it was "easy" to trigger nuclear spin isomers. However, understanding of the reaction pathways involved required more precise tools for triggering and attention turned to the use of synchrotron radiation (SR) sources of tunable monochromatic X-rays.

This final report describes work continued with AFOSR support using SR sources for triggering the release of the energy stored in the ^{178}Hf isomer during the period March 1, 2001 to August 31, 2005. However for continuity, it is convenient to recall that positive confirmation and extension of those first results were subsequently reported with a nearly annual frequency [2-8]. Interleaved with those publications were reports of experiments purported to disprove all of the positive results [9-12]. Such disagreements seemed to arise from the difficulties in mastering this application of the complex techniques of nuclear resonance spectroscopy to further excite already excited states of nuclei present in physical samples only in very low concentrations. However, recent publications outside of the professional journals of some sensational perceptions of societal implications of the triggering of isomeric nuclei has created a political climate that hindered efforts to focus more closely upon an understanding of the Physics of the process [13]. Moreover, the paucity of available SR sources for such work, together with the long lead times in gaining access to them, has made progress up the learning curve rather stochastic.

During the most recent period covered by this report, emphasis was turned to the simplest possible demonstration of triggering of the 31-yr isomer of ^{178}Hf. The energetics of the excitation

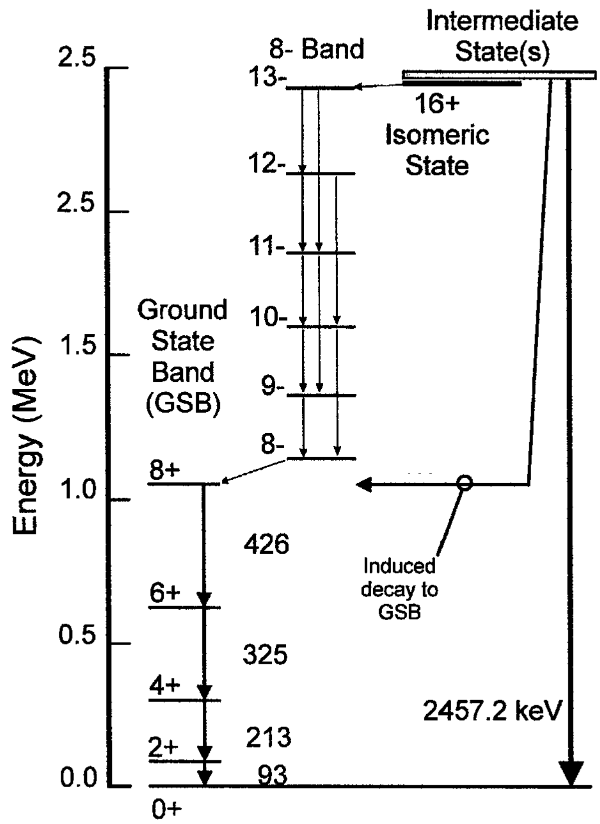

Figure 1 Schematic diagram of the energies of a few of the excited states of the ^{178}Hf nucleus. Total spin and parity of each state is shown to the left of each level. Transitions within the Ground State Band (GSB) are shown by the heavy arrows with energies of the corresponding gamma-photons emitted to the right. Smaller arrows show transitions leading to the GSB that result from spontaneous decay of the 16+ isomeric state. The absorption of an X-ray photon induces decay of the 16+ state by raising it to the intermediate state that decays through one of several branches.

scheme is shown in Fig. 1. The long lifetime of the initial 16+ isomeric nuclear state results from an alignment of the spins of four of the constituent particles of the nucleus. Such alignment of four particles is reflected in the nomenclature describing it as a "4-QP" isomer. A shorter-lived "2-QP" isomeric state in the same nuclide results from the alignment of the spins of only two constituents.

Selection rules upon changes of angular momentum and upon changes of its projection upon the figure axis of the ^{178}Hf nucleus forbid the electromagnetic transitions that would otherwise allow spontaneous emission of cascades of gamma-photons to radiate away the excitation energy. However, as we proposed in 1982 [14], an isomeric state might be able to absorb an X-ray photon and make a transition to an excited state of greater energy that would be better able to subsequently radiate away its excitation energy. Such an intermediate state would have to have a "mixed character" [2-7] so that the selection rules upon changes of its angular momentum and spin would be relaxed. It would be reasonable to expect that the output transitions from the intermediate state would have branches into several possible cascades of gamma emissions, necessarily including one branch back to the initial isomeric state. In the final work reported here another one of the output branches has been found that consists of a single transition to the ground state as shown in Fig. 1. Such a favorable arrangement of branches facilitated our determination of the energy of one intermediate state to be 2457.20(22) keV. This means that direct absorption of an X-ray photon of 11.15(27) keV can release the 2446.06 keV stored by a ^{178}Hf isomeric nucleus, without electron involvement to conserve energy [15].

All of the achievements realized under this AFOSR Grant have been published in readily accessible journals. The details of the experimental method and techniques of data analysis have been fully described in a PhD dissertation, copies of which are available from UMI dissertations [16].

Confidence in the positive results is very high and the status at the time of writing is summarized in an open letter reproduced in the Appendix. Selected results from the published works that illuminate particular challenges posed by the negative results obtained elsewhere are reviewed in the following section.

ACCOMPLISHMENTS - Critical Issues Resolved by this Research

Size of the Effect From the first report of the triggering of the decay of the ^{178}Hf isomer, one of the disturbing concerns was the unexpected (and favorable) size of the effect. A large integrated cross section was found that meant relatively small samples could be considered for "ignition" in practical applications in which self-sustaining chain-reactions of energy release could be initiated with a relatively small trigger energies. The first experiments had used a broadband source of pulsed X-rays and only integrated cross sections could be derived from the data. Values were large and corresponded to the Breit-Wigner limiting cross section in "depth" and to about 1 eV in width [1,2]. While this was an unprecedented size, contrary to critical opinion, it did not violate any fundamental limits nor require any "new Physics." Subsequent experiments with monochromatic SR X-rays revealed the source of the large effect.

Excitation Spectroscopy Consistently repeated experiments at synchrotrons in Japan and Switzerland proved that the excitation function for the triggering was structured and was peaked near critical spectroscopy "edges" for photoionization of the inner shell electrons that surrounded the isomeric nuclei in the physical sample [5]. Typical data are shown in Fig. 2 and Fig. 3, below, reproduced from publications [6,7,15].

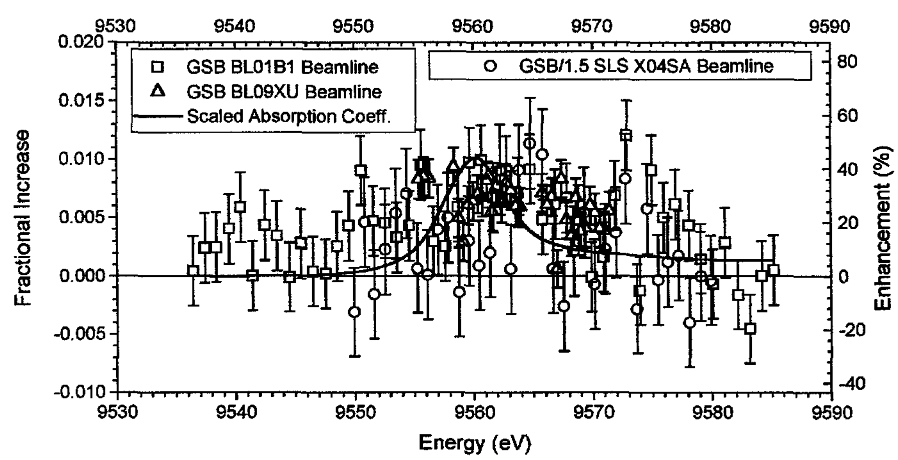

Figure 2 Fractional increases in the number of gamma-photons collected in the GSB of ^{178}Hf together with the relative absorption coefficient scaled to fit the vertical range to show the Hf(L3) photoionization edge. Data from three beamlines are shown as marked. The scale to the right has been corrected for the duty cycle of the SR excitation. In the legend, SLS identifies data taken at the Swiss Light Source. Other beamline designations refer to SPring-8.

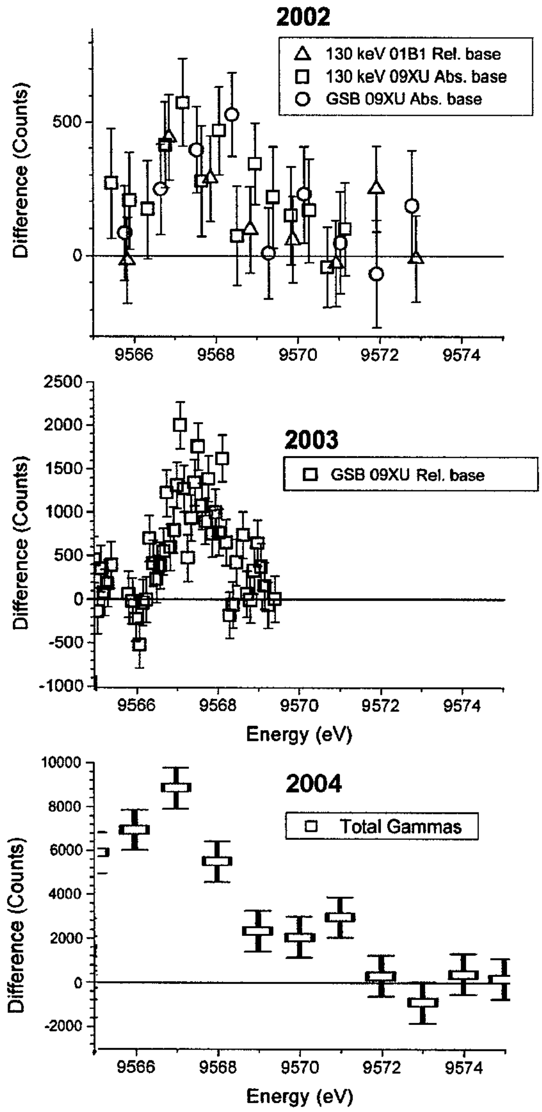

Figure 3 Differences in the numbers of gamma photons detected in several different nuclear transitions populated when ^{178}Hf isomers were triggered by pulses of monochromatic SR X-rays at the energies shown on the abscissae. Data shown here were obtained at the SPring-8 SR source and are reproduced from publications [6,7,15] that completely describe how the data were taken and analyzed. The term GSB refers to the sum of photons detected in transitions in the ground state band of the ^{178}Hf nuclide and the designation 130 keV refers to a transition excited only by triggered decay of the isomer. "Abs." and "Rel." base refer to increases in the number of photons detected above what were obtained from spontaneous decay of the isomeric nuclei in the sample when the irradiating X-ray beam was blocked and when it was set to an X-ray energy believed to be unable to trigger the decay of the isomers. Though not so marked, the 2004 data are increases above an "Abs." base reference and were visible in "real time" on a monitor as the X-ray energies were stepped to different values. Significant is that the peak effect occurs ~6 eV in X-ray energy above the Hf(L3) photoionization edge at 9561 eV for the electrons, while the details of the fine structure between the edge and the peak seem to depend upon sample preparation and history of usage.

Calibrated comparisons between the functional curve for yield and the functional curve for photoionization showed that the probability for triggered decay of the ^{178}Hf isomer was 0.16% of the probability for photoionization of an Hf(L3) electron in the sample [6]. In a sense this explains the unexpectedly large size of the cross section for triggering energy release from the ^{178}Hf isomer. Inner shell electrons help mediate the transition to the state reached by absorption of a trigger X-ray. That intermediate trigger level must be a mixed state involving excited inner shell electrons as well as excited nuclear states, much as in the well-known case of internal conversion of nuclear excitation.

Induced Paths of Decay Absorption of a monochromatic X-ray by the 16+ isomeric state shown in Fig.1 triggers the decay of the state by raising it in energy to a more unstable intermediate ("trigger") level. Controversy arose from the unjustified original expectations that in the unlikely event that the 16+ isomer could be triggered to decay, then "surely there could be only one way in which that could happen." This *a priori* fallacy was disproved as described in publications [15, 17] and 3 observed types of induced decay are shown in Fig. 1. The one leading first to members of the 8- band follows part of the gamma cascade seen in the spontaneous decay and is characterized by the emission of a well-known spectrum of gamma rays that includes a component transition at 495.5 keV. Because the first cascade through the 8- band leads to a 2-QP isomer with a half life of 4 seconds, decay through that channel populates the subsequent members of the GSB only after a statistical time lag of ~4 s.

The transition at 495.5 keV is of no particular significance other than it is one of several to be immediately produced after onset of a spontaneous decay event and it was the strongest transition we reported in the first description of the triggered decay of the ^{178}Hf isomer [1]. That first experiment used broadband X-rays. Criticism of subsequent work persisted [18] because enhanced emission of 495.5 keV gammas was not reported afterwards, but then those subsequent experiments used monochromatic X-rays at energies concentrated around the Hf(L3) photoionization edge. It was reasonable to believe that the decay cascade of gamma photons that included those at 495.5 keV were triggered at some X-ray energies present in the broadband source and not in the monochromatic SR X-rays used in subsequent work. That proved to be the case as described in detail in the publication [17]. Figure 4 shows triggered emission of the 495.5 keV line, together with some other transitions as originally described [1].

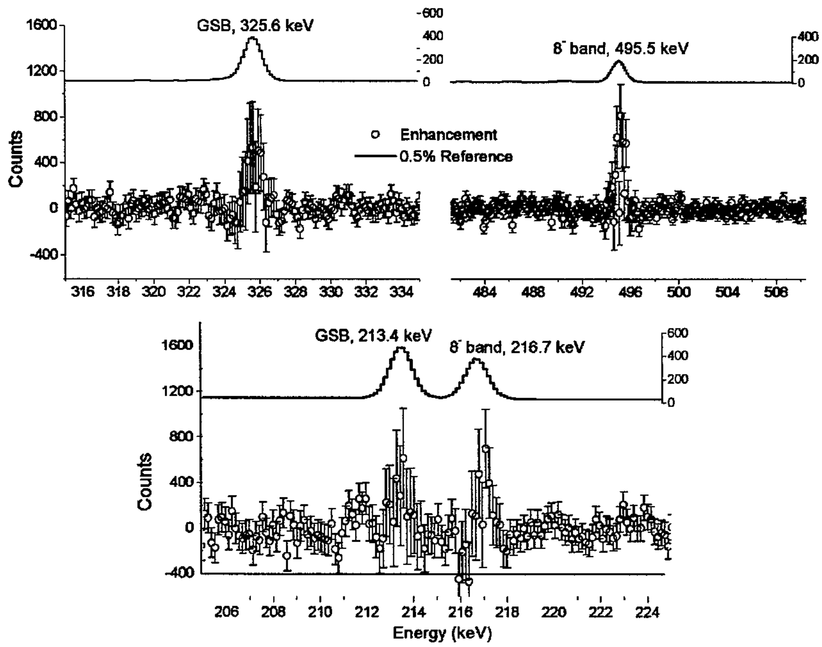

Figure 4 Increased numbers of gamma photons detected in members of the 8- decay cascade at 216.7 keV and 495.5 keV together with members of the GSB at 213.4 keV and 325.6 keV that were triggered by irradiation with monochromatic SR X-rays at energies near 20,825 eV.

By gating the beam of SR X-rays on a scale of seconds it was found that the members of the GSB were detected after a statistical time lag of 4 seconds with respect to the times the members of the 8- band were detected [17], as was expected from Fig.1. The excitation function for triggering decay of the isomer through the 8- band is shown in Fig. 5, reproduced for convenience from the publication describing methods,

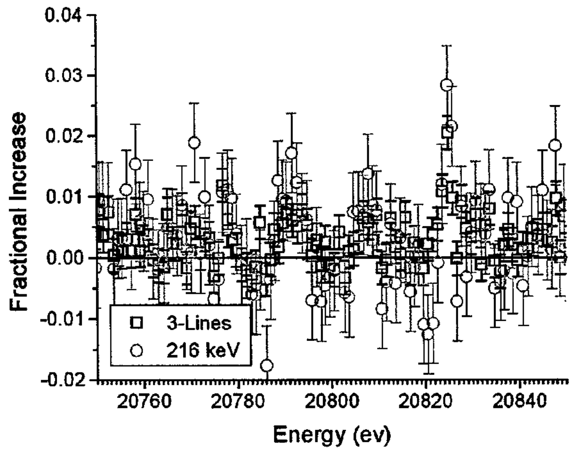

Figure 5 Excitation function for inducing decay of the ^{178}Hf isomer through a member of the 8- band at 216.7 keV together with that for inducing three members of the GSB using monochromatic SR X-rays at SPring-8.

data, and analyses [17].

For the other two decay cascades shown in Fig. 1 that bypass the statistical time lag associated with the 8- band head, "new" gamma transitions not observed in spontaneous decay were expected and were reported. The most complete description is found in the Rusu PhD Dissertation [19] that became a benchmark for "proof" of the efficiency for triggering the decay of the ^{178}Hf isomer. Much of that landmark work was done with coincident detection of "new" lines and known members of the GSB. That, too, attracted strident criticism since the methods of measurement and analysis were complex. There was persistent demand for a demonstration of the emission of gamma photons from a line not present in spontaneous decay of the isomer and that was pronounced enough to be seen in direct, simple single photon detection. Figure 6 reproduced from publications [7] shows such a feature.

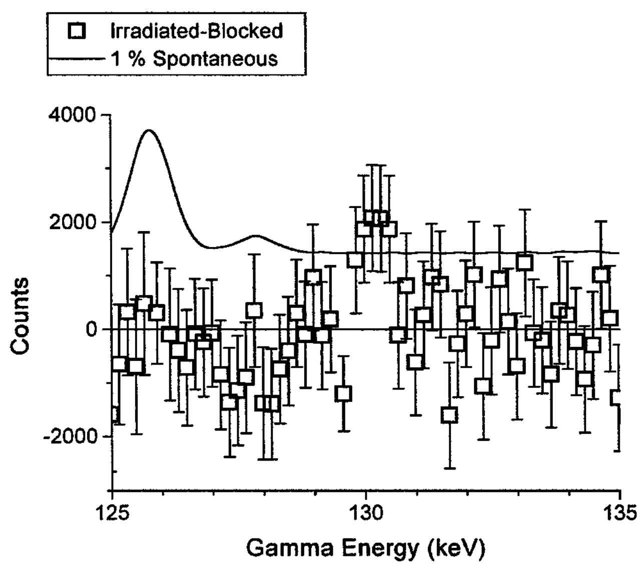

Figure 6 Differences in the numbers of gamma photons detected from a sample of ^{178}Hf isomers with and without monochromatic SR X-ray irradiation together with 1% of the numbers of gammas detected from spontaneous decay in the sample for scale. The peak showing triggered excitation of a "new" transition at 130 keV corresponds to the feature studied in coincidence in the Rusu Dissertation [19].

Immediate Triggered Decay Published results [6,7] of experiments performed at the Swiss Light Source (SLS) at Villigen benefitted from instrumentation that could record the passage of single bunches circulating in the synchrotron ring. Moreover preselected patterns of electron bunches could be injected into the ring so that the effects of irradiation occurring when the bunches created SR X-rays could be compared with spontaneous emission from an isomeric target when there were deliberately introduced vacancies in the bunch pattern. The temporal pattern of irradiation correlated with the circulating of the electron bunches and vacancies and could be detected with an avalanche photo diode (APD). Figure 7 reproduces data from publications [6,7].

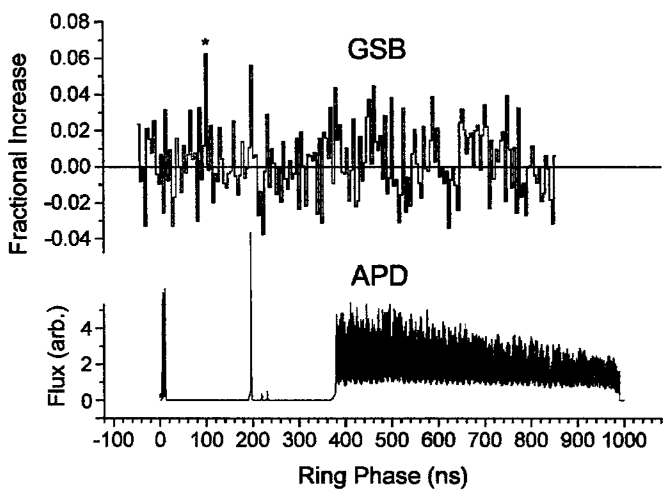

Figure 7 Fractional increase in the number of gamma photons detected in transitions of the GSB as a function of the phase of the passage of the pattern of electron bunches circulating around the storage ring. Transit time around the ring was nominally 1000 ns, so the figure shows the accumulation of repeated cycles of passage of the bunches. The structure marked with the * was an artifact that produced increased counting rates at all gamma energies for that one phase.

The APD device could not resolve the actual passage of a bunch which is typically about 20 ps in duration, but rather averaged that passage over the 2 ns separation between bunches. The X-rays were emitted only during the 20 ps of actual "current" corresponding to the passage so the duty cycle for triggering was only about 1% (*ie.* 20 ps of electrical current producing X-rays followed by 2 ns of no current.) However, the spontaneous emission of gamma photons from the sample was continuous. So a 4% average increase such as indicated in Fig. 7 that occurred entirely during the 1 % of the time there was current making X-rays actually corresponded to a 400% increase in gamma ray emission during the time the SR X-rays were "on."

Alignment and Confidence One of the larger challenges in collecting reproducible data were the stochastic sources of "noise." The SR operator often realigned the bunch path at discretion and other sources of mechanical and electrical shifts were frequent. To achieve the objective of a simplest possible proof of triggering a real-time diagnostic was developed. All details are described in the Zoita PhD Dissertation [16] and are summarized in considerable detail in the most recent publication [15]. With proper care of the sample and alignment of it in the SR X-ray beam, the collection rate of the total numbers of gamma photons of all energies (above 100 keV) could be "seen" in real time to increase with triggering. This provided a significant indicator for aligning experiments by optimizing the observed rate of gamma counting by "tuning" the physical position of the target in the beam and similarly by tuning the energies of the monochromatic X-rays. Figure 8 reproduces [15] a typical measurement of the total numbers of gamma photons collected as the sample was remotely moved through the monochromatic X-ray beam by a stepping motor.

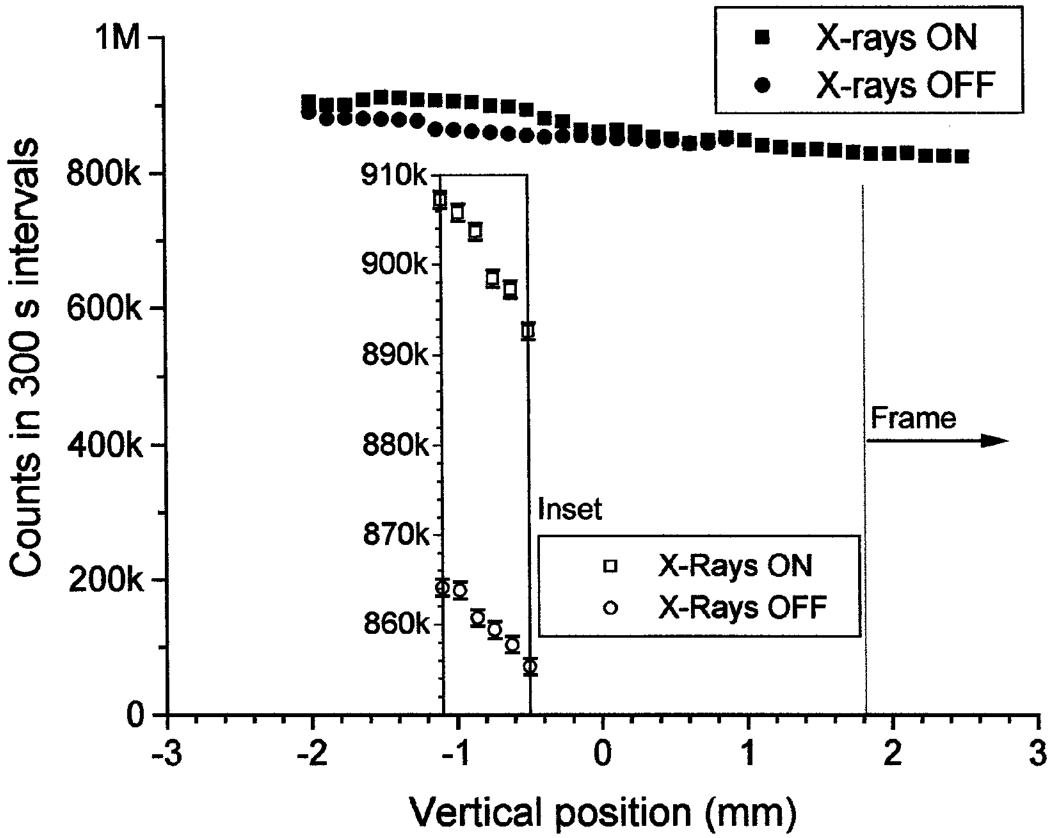

Figure 8 Numbers of gamma photons of all energies above 100 keV collected as the physical sample of isomers was moved through the profile of the SR beam of monochromatic X-rays. The inset shows a magnification enabling the error bars to be seen. Notable is that even when the beam of X-rays was stepped onto the massive frame holding the sample, no scattering or other anomalies were detected.

The confidence that there was a total increase of the number of gamma photons triggered by the passage of the sample through the monochromatic beam of SR X-rays was very large, being 92 sigma in statistical terms [15].

SIGNIFICANCE OF THE ACCOMPLISHMENTS

In the nomenclature in which "four 9's" of confidence would represent 99.99%; the confidence proven by statistics of 92 sigma is 1,824 9's. Hafnium isomer triggering has been proven. A comparison of the high confidences for positive experiments and low confidences of negative experiments is summarized in Appendix 1.

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INTERACTIONS/TRANSITIONS

Under this Grant, interactions have been intense, but informal. The most notable level has been sustained with colleagues at Sandia, in particular Dr. Pat McDaniel with whom frequent consultations continue.

A major transition was achieved because of interest at DARPA. In 2003, much of the project was transferred to DARPA with significant increase in funding to facilitate the pace of further research into nuclear spin isomers.

PERSONNEL

Professional personnel supported and/ or associated with this project have been:

1) Carl B. Collins	UTD Professor and Principal Investigator
2) Farzin Davanloo	UTD Research Scientist
3) C. Rusu	UTD Graduate Student
4) C. Zoita	UTD Graduate Student
5) C. A. Ur	Research Scientist, HHNIP&NE Bucharest, Romania

6) I. I. Popescu	Academician, IGE Foundation, Bucharest, Romania
7) V. I. Kirischuk	Research Scientist, GKTCenter for Nuclear Materials, Kiev, Ukraine
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9) J. M. Pouvesle	Research Director, GREMI, Univ. d' Orleans, France
10) S. Emura	ISIR, University of Osaka, Japan
11) T. Uruga	JASRI, Spring-8, Staff Scientist, Hyogo, Japan
12) Y. Yoda	JASRI, Spring-8, Staff Scientist, Hyogo, Japan

Appendix I

Reprint of the Letter
being circulated upon request

Summarizing the confidence of all Hf-isomer triggering experiments

by C. B. Collins

UT|D

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"I thought I had put the hafnium bomb where it belongs .... We killed it with laughter." – Peter Zimmerman, p.49 *Physics Today*, Jan. 2005.

Summary of the Confidence Levels Supporting Hf-Isomer Triggering Claims in 2005

The stimulated isomer energy release (SIER) from the 31-year isomer of Hafnium-178 was first proven at Orsay, France on March 24, 1996 with the inelastic scattering of alpha particles from a unique isomer target that had been produced at Dubna in Russia. The yield was over 300 times the recommended upper limits (RUL's) that define the largest possible efficiencies for nuclear reactions occurring by known mechanisms. For almost a decade since, those results have been confirmed and reconfirmed by triggering with broadband X-rays from Bremsstrahlung devices and from monochromatic X-rays from synchrotron radiation (SR) sources by an international team of 15 scientists from 7 laboratories in 6 countries. All confirmations and reconfirmations have been subject to peer review and have been published. The significance is real; and it is proven.

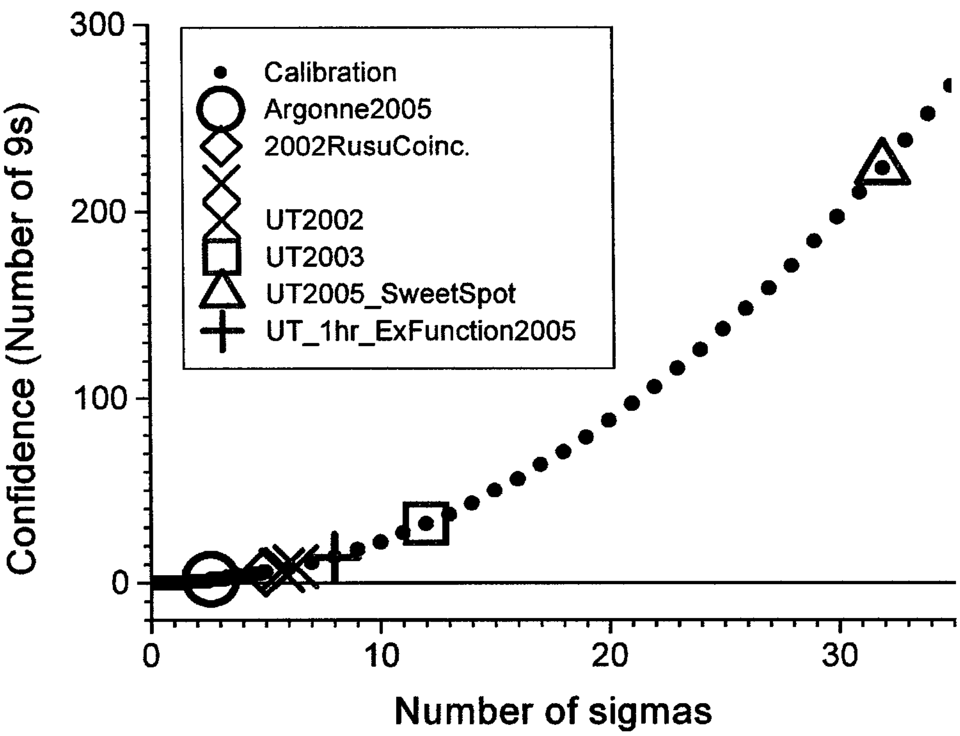
A "controversy" has been manufactured by a few scientists, some quite prestigious; and their arguments have depended upon uncertainty in what constitutes "proof." They have been sustained further by political force. Certainly, some researchers have seen nothing in experiments purported to be testing for isomer triggering. That is the normal situation in a rapidly developing field of research. Some results are positive; some are negative. However the measures of confidence for the positive results are far greater than the confidence limits for the negative conclusions.

The following graph summarizes the levels of confidence for the recent results. Confusion often results from the statement for confidence levels in terms of "sigmas." Sigmas are the "standard deviations" of the counting statistics for the photons produced by SIER. The black points show the mathematical connection between "sigmas" and the number of 9's of confidence. For example two nines would be 99% confidence, while six nines would be 99.9999% confidence. This is an established mathematical relationship between confidence and sigmas.

Confidence reported by the authors for the negative results obtained at Argonne [1] over 3 years of performance is shown by the large black open zero. Coincidence measurements are plotted in blue. Positive results for the 2002 Rusu Dissertation results [2] upon which TRIP was modeled are shown by the diamond symbol. ***Confidence for the positive coincidence measurement is about 1000 times greater than for the negative results of Ref. [1].*** The positive measurements from the Texas Group are shown in red and are typified by the results from Fig.5 of Ref. [3] that are shown here by the red "+" symbol read on the left scale. Confidence is fourteen nines, meaning certainty of 99.999999999999%. The "best" positive results, reported in

Table 1 of Ref. [3], would plot off the scale of abscissas. Confidence is 1,824 nines – far too many to write out explicitly

It is incorrect to assert that "Hf-isomer triggering has not been proven." Such a statement is simply politics. It is correct to say that for whatever reason, such triggering has not been universally accepted.



[1] I. Ahmad, et al., Phys. Rev. C71, 024311 (2005).

[2] C. Rusu (PhD Dissertation, U of Texas at Dallas, 2002)
Available from: http://tls.il.proquest.com/umi/dissertations/disexpress.shtml
(Order Number: 3087127).

[3] C.B. Collins, et al., Laser Phys. Lett. **2**, 167 (2005).

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