Stewardship Science Academic Alliances Program

Nuclear Stewardship Research.

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Introduction

The second year of our research program has been marked by significant success and progress. It has also been marked by significant changes both in the personnel and location of the major experimental research program. This report covers the period roughly from August 2004 through May 2005. During this period our research has focused mainly on applying the surrogate reaction technique and the 'ratio' method to deduce neutron induced fission cross sections on uranium nuclei.

The following lists (in more or less chronological order) some of the highlights of the program during 2004-05.

- The first production experiments, to investigate the ^{236,238}U(d,pf) and ^{236,238}U(d,d`f) reactions as surrogate for the neutron induced fission reactions ^{236,238}U(n,f) and ^{235,237}U(n,f), respectively, were carried out at Yale.
- Progress reports and results from these experiments were presented by Cristina Plettner (Yale), Con Beausang (Yale and Richmond), John Ai (Yale), Jennifer Church (LLNL) and Lee Bernstein (LLNL) at a variety of international conferences and workshops including:
    - Nuclear Reactions on Unstable Nuclei and the Surrogate Reaction Technique Workshop, Asilomar CA., January 2004.
    - The 2004 SSAA Symposium, Albuquerque N.M., March 2004.
    - The RIA Summer School, Argonne National Laboratory, August 2004.
    - ND2004: Nuclear Data for Science and Technology, Santa Fe, N.M. October 2004.

- The Division of Nuclear Physics, American Physical Society meeting, Chicago, IL., October 2004
  - The NUSTAR Conference, University of Surrey, U.K., January 2005.
  - The Yale Workshop on Nuclear Structure Physics Near the Coulomb Barrier, June 2005.
- Our results include (importantly) benchmarking the surrogate method by using the ^{236}U(d,pf) and ^{238}U(d,pf) reactions as surrogates for the ^{236}U(n,f) and ^{238}U(n,f) reactions, respectively.
- Importantly, we reported the first surrogate measurement of neutron induced fission on ^{237}U, via the ^{238}U(d,d'f) reaction, up to equivalent neutron energies of ~14 MeV.
- Manuscripts detailing these results have been accepted for publication in Phys. Rev. C, Rapid Communications, and in J. Phys. G. [1,2]
- We have developed a new and potentially very powerful method of reporting the results of our surrogate measurements as ratios of cross sections. This has the effect of removing many of the systematic errors associated with the absolute cross section measurement [1,2].
- In December 2004 and March and May 2005 we collaborated on experiments at LBNL, using the STARS and Liberace Arrays to investigate the ^{236}U(α,α`f) and $^{238}$U($\alpha$,$\alpha$`f) and ^{236}U(^{7}Li,xf) and ^{238}U(^{7}Li,xf) reactions as surrogates for the ^{235}U(n,f) and ^{237}U(n,f). In addition to much improved statistical accuracy compared to the first Yale experiments, these data provide important complementary information to allow us to check and compare the validity of the surrogate approach by measuring with a variety of different reactions.
- The analysis of these data sets will form the thesis of John (Ho-Chaing) Ai, the graduate student supported by the SSAA project.
- Regular visits and conference calls continue to coordinate the program and collaboration.

The last year also saw significant changes in the personnel and in the location of the primary experimental program

- In August 2004 Professor Beausang relocated to the University of Richmond to take up his new appointment as Associate Professor and Robert E. and Lena F. Loving Chair.
- In March 2005, our postdoctoral fellow, Dr. Cristina Plettner, left to assume a permanent position as a research scientist with the IBM Corporation. Dr. Plettner duties at IBM include responsibilities for experiments detailing radiation damage effects on chips.
- We are actively searching for a replacement for Dr. Plettner.
- Following the move of Dr. Beausang to the University of Richmond, the primary location for the experimental program utilizing the STARS detectors has shifted to the 88-Inch Cyclotron at LBNL. While experiments are still envisioned for Yale it is likely that the majority of the experimental program in the remainder of 2005 will be carried out at LBNL.

Despite these changes in personnel, the second year of the program saw significant sustained progress and the publication of significant new results. In short, we have already achieved and surpassed many of our planned major milestones for the project. Additional details on some of these highlights are presented below as well as some of our plans for future research directions for the coming year or so.

Key Personnel

		% Support from the Grant
Yale University		
Cornelius W. Beausang,	Associate Professor, P.I.	0
Cristina Plettner,	Postdoctoral Fellow	100
Ho-Chaing Ai,	Graduate Student	100

In addition, the following persons from Yale University and from LLNL have participated in experiments or played a significant role in the commissioning of the STARS detector at Yale.

Yale University

R.F. Casten	Professor
A. Heinz	Assistant Professor
D.A. Meyer	Graduate Student
E.A. McCutchan	Graduate Student
J. Qian	Graduate Student
G. Gurdal	Graduate Student (Clark University)
E. Williams	Graduate Student
B. Crier	Undergraduate Student (University of Richmond)
J. Ashenfelter	Director of Operations, WNSL
T. Barker	Electronics Engineer, WNSL
W. Garner	Vacuum Specialist, WNSL
R. Wagner	Electronics Technician, WNSL

Lawrence Livermore National Laboratory

Lee Bernstein	Staff
Larry Ahle	Staff
Jennifer Church	Postdoctoral Fellow
Jason Burke	Postdoctoral Fellow

Summary July 2004 - May 2005

The second year of our Academic Alliances Stockpile Stewardship research program continued the progress made in year one. Despite the relocation of the P.I. in August 2004, the program has continued with regular collaboration meetings, successful joint experiments (with the LLNL and LBL groups) at the 88-Inch Cyclotron in December 2004, and March and May 2005, the submission or publication of several articles, including a

Phys. Rev. C. Rapid Communication, and the presentation of several invited talks at international meetings.

3.4.b. The Surrogate & Ratio Methods and results for the actinides:

The ^{236,238}U(d,d'f) and ^{236,238}(d,pf) reactions at deuteron beam energies of 24 and 32 MeV were investigated using STARS and YRAST ball at Yale over a six week period during April and May 2004. These reactions serve as surrogates [3,4,5] for neutron induced fission on ^{235}U / ^{237}U and ^{236}U / ^{238}U, respectively.

While reactions on ^{235}U are very well studied, because of its short half life essentially no information was previously available for ^{237}U.

Importantly, our results for the ^{236,238}U(d,p) reactions can be compared to the well known results for ^{236,238}U(n,f) and hence can benchmark the method. The reactions utilized to date are summarized in Table 1.

Surrogate Reaction	Beam Energy	n-induced reaction of Interest	Equivalent neutron energy	
^{236}U(d,d'f)	24 and 33 MeV	^{235}U(n,f)	Up to 14 MeV	Benchmark
^{236}U(d,pf)		^{236}U(n,f)		Benchmark
^{238}U(d,d'f)		^{237}U(n,f)		New
^{238}U(d,pf)		^{238}U(n,f)		Benchmark
^{236}U(α,α'f)	55 MeV	^{235}U(n,f)		Benchmark
^{238}U(α,α'f)		^{237}U(n,f)		New

Table 1: Reactions studied using STARS.

The application of the surrogate technique however may have several limitations and certainly poses certain questions. For example, does the light ion induced reaction produce a compound nucleus? What are the direct reaction contributions and how do they differ from neutron induced reactions to reactions induced by other light ions? What is the correct/best model to use to calculate $\sigma_{CN}(E)$, which presumably should be written $\sigma_{CN}(E,I)$, where I is the angular momentum. What are the effects of the different angular momentum distributions resulting from the light-ion versus neutron induced reactions on the decay probability?

In addition to these 'physics' issues there are often additional practical limitations associated with the determination of the decay probability. For example, considering our surrogate (n,f) measurements in uranium nuclei;

$$P_{(n,f)}(E) = N(d,pf) / N(d,p).$$

Here N(d,p) is the total number of (d,p) events while N(d,pf) is the number of these events which lead to decay via (for example) the fission branch. Due to practical issues involving target construction and contamination and to additional (random) contributions from deuteron breakup reactions, determining N(d,p), the denominator in the above equation, can impose serious systematic and statistical limitations on the technique.

To bypass these, in our recent experiments we instead present the results as ratios of cross sections. In other words we report,

$$\frac{\sigma(n,f)(^{236}U)(E_x)}{\sigma(n,f)(^{238}U)(E_x)} = \frac{P(d,pf)(^{236}U)(E_x)}{P(d,pf)(^{238}U)(E_x)} \approx \frac{N(d,pf)(^{236}U)(E_x)N_R(^{238}U)}{N(d,pf)(^{238}U)(E_x)N_R(^{236}U)}.$$

In this equation, N_R (the number of Rutherford scattered particles) is an easily determined normalization factor required to account for different target thicknesses and beam fluxes.

This experimental ratio, extracted from the Yale data, is presented in figure 1 for both the (d,d'f) reaction as well as (and perhaps more importantly) for the (d,pf) reaction on both ^{236}U and ^{238}U targets. It is important to note that the (d,pf) reaction ratio is surrogate for the well known ^{236}U(n,f) / ^{238}U(n,f) ratio and serves to benchmark both the surrogate method and also the ratio method. A typical spectrum showing candidate (d,d'f) fission events from which this ratio was obtained is shown in figure 2.

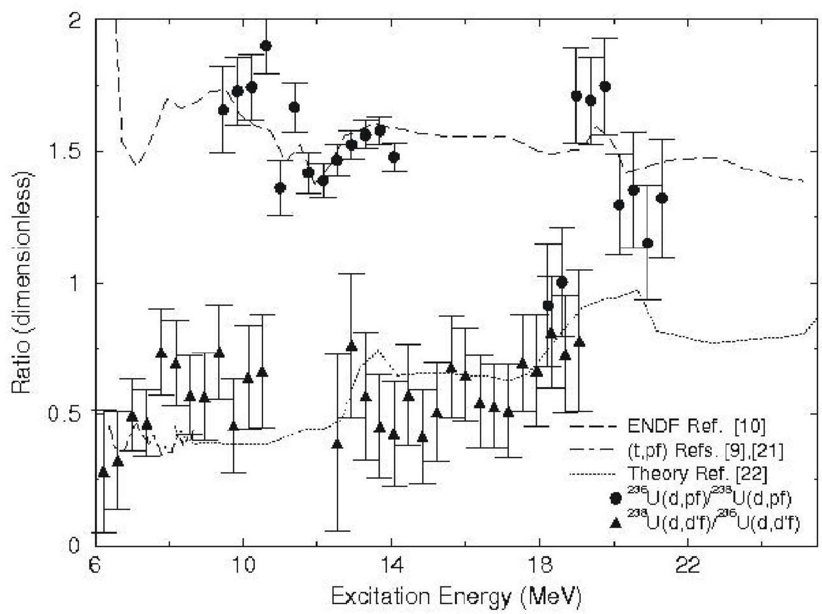

Figure 1: Ratio of cross sections for both the ^{236}U(d,pf)/^{238}U(d,pf) (circles) and ^{236}U(d,d'f)/^{238}U (d,d'f) (triangles) extracted from our data and plotted as a function of excitation energy. The experimental ratios are compared to both preexisting data [6,7] and to the results of modern calculations [8,9].

As can be seen in figure 1, our experimental ratio for the (d,p) reactions is in good agreement with the ratio extracted from directly measured data and with the calculated values and trends.

Therefore, we can move forward (with some confidence) and also apply the ratio method for the (d,d'f) reactions. In this case, although the error bars are larger, the results show an impressive agreement with the calculations and, where available, with the existing data. In fact, we can then use the known cross sections for ^{235}U(n,f) to deduce, for the first time the ^{237}U(n,f) cross section up to an equivalent neutron energy of ~ 14 MeV. Manuscripts detailing these results have been accepted in for publication in Phys. Rev. C. and in J. Phys. G. [1,2].

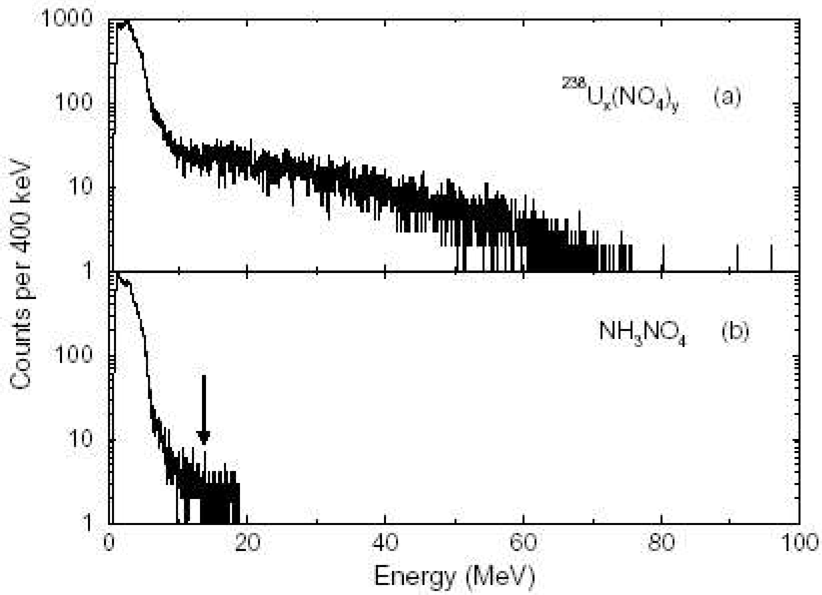

Figure 2. Spectra showing a) candidate fission events following the ^{238}U(d,d'f) reaction and b) a background 'fission' spectrum extracted using an identical sort procedure from data obtained following (d,d') reactions on a NH_3NO_4 target. Any contamination in the top spectrum stops at energies less than ~14 MeV.

Recently, the ^{236}U(α,α`f) & $^{238}$(α,α`f) reactions were investigated using the STARS and LIBERACE Arrays at LBNL (during December 2004 and March 2005). Preliminary analysis of these data reveal that the statistics are much improved over the first Yale experiments and the extracted ratios, shown in figure 3 for the ^{236}U(α,α`f)/$^{238}$(α,α`f) ratio, are generally in agreement with the Yale (d,d') data and with the theoretical calculations.

The ratio method opens up a wealth of possibilities and opportunities as well as several interesting questions to be addressed in our future research. For example, can the ratio method be applied to other mass regions? How similar do the structures of the nuclei involved in the ratio have to be in order for their individual structure affects to cancel in the ratio? These and other questions will be addressed over the remainder of the current proposal by continuing analysis of our existing data sets and by additional experiments.

Data analysis is continuing at LLNL, Richmond and Yale. In particular, the target contamination issue which plagued the (d,d') data sets has been mostly resolved in the LBNL data. Hence these data provide an excellent opportunity to extract the absolute cross section and perform a true surrogate measurement using this data. This analysis will form part of the thesis project of John (Ho-Chaing) Ai at Yale. So far the results look promising.

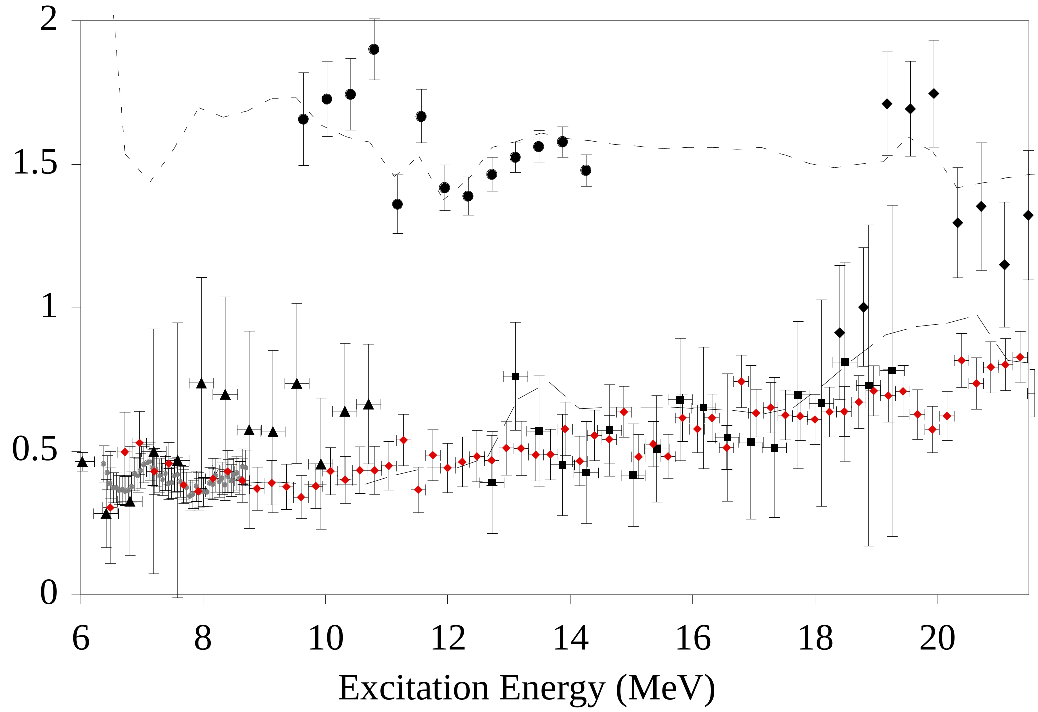

Figure 3 Preliminary ratio of cross sections (lower ratio, red circles) for the $^{236}U(\alpha,\alpha`f)/^{238}(\alpha,\alpha`f)$ extracted from the STARS/LIBERACE data set.

Outlook for 2005-06

The outlook for the coming twelve month grant period looks extremely promising.

In addition to the ratio method for presenting our results, a second novel and potentially useful and exciting idea has come from these recent experiments. The idea is the following: Let us use the gamma-ray detectors, YRAST Ball or LIBERACE (or our proposed dedicated sum-energy multiplicity filter), to detect fission events rather than using the STARS SiLi detectors directly.

The gamma-ray multiplicity for actinide nuclei, populated via such light ion reactions, and which do not fission, is very low, probably less than 1 – 2 on average. This is due both to the small angular momentum input from the reaction and to the often very large internal conversion coefficients associated with the low energy transitions depopulating the lower spin states (this is particularly the case for even-even nuclei). In contrast, the gamma-ray multiplicity associated with fission fragments can be quite high, on the order of 5 – 10 transitions per fragment or a total gamma-ray multiplicity of 10 – 20. Thus detection of a gamma-ray, certainly a gamma-ray with energy greater than ~100-200 keV (above both the U x-rays and the energies of most low-spin yrast transitions) is should be essentially equivalent to, and independent of, detecting the fission fragment directly.

If this idea works, then even with modest gamma-ray detection probabilities the efficiency for ‘fission detection’ can be quite high. For example, the total photopeak efficiency of the YRAST ball array is on the order of ~3% for ~ 1 MeV gamma-rays. However, what matters in the current discussion is the total gamma-ray detection efficiency (counting both photopeak and Compton events) and this is at least 6%. Therefore, for multiplicity $M_\gamma = 10$, the probability of detecting anyone of the gamma-rays is on the order of 60%, or more than twice as large as the geometric efficiency of the STARS SiLi detector.

A comparison of the yield of fission fragments and gamma-rays plotted as a function of scattered alpha-particle energy is shown in figure 4. As can be seen the statistics in the gamma-ray yield spectrum are larger than those in the fission spectrum and the general trends in the two spectra are the same for low to medium excitation energies. Significant deviations are observed in the two yield spectra for higher excitation energies and the sources of these are currently being investigated. It is clear however, even from these preliminary sorts, that the method shows promise.

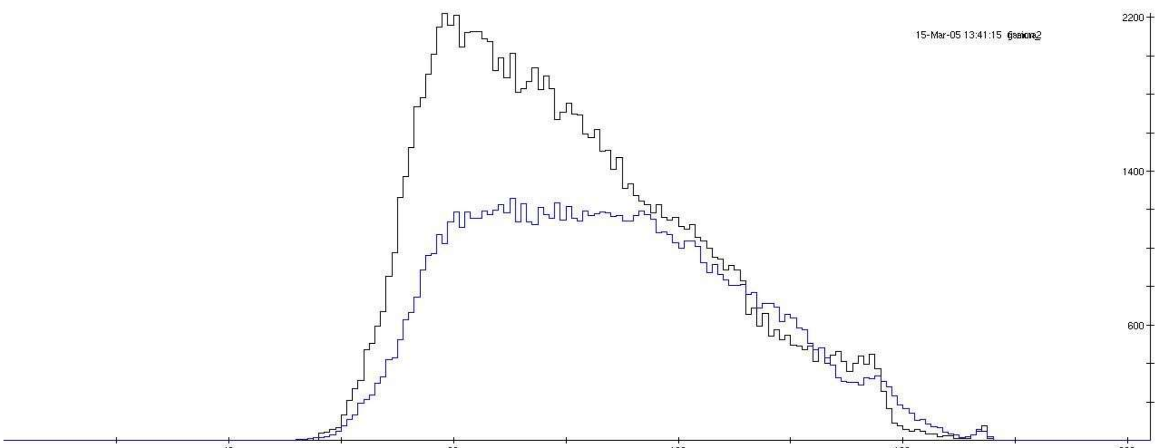

Figure 4: Comparison of fission yields measured via direct detection of the fission fragment (top spectrum) or via gamma-ray yield (lower spectrum) plotted as a function of scattered alpha-particle energy.

It is important to note that one does not need to measure the gamma-ray energy or to identify which fragments are involved (although, clearly, this might provide important additional information on the peak mass yields). Thus, very large detection efficiencies could be achieved at a very modest cost by constructing a small compact BGO (scintillator) array surrounding the target. Such an array could accommodate the high resolution clover detectors of YRAST Ball or LIBERACE and could easily have efficiencies approaching 90% or more for detection of high multiplicity gamma-ray cascades. The gamma-ray multiplicity from direct reactions of light ion contaminants in the target (typically oxygen, carbon or nitrogen) should also have low multiplicity. In our (pending) grant renewal application, in addition to continuing out physics program on the surrogate technique, we propose to construct such a compact sum-energy multiplicity filter for use with the STARS detectors. Such a device could be particularly useful when attempting experiments with very short half-life isotopes where the target based alpha activity may well compromise the STARS SiLi detectors, but would not affect the gamma-detectors.

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Publications and Invited and Contributed Talks

Stewardship

STARS experiments on actinides--preliminary results.
C. Plettner, et al., RIA Summer School, Argonne National Laboratory, July 2004

Measuring reaction rates with STARS -- ^{92}Zr(d,d) reaction
H. Chiang-Ai, et al., RIA Summer School, Argonne National Laboratory, July 2004, Chicago.

Fission yields for ^{236,238}U(d,df)
C. Plettner, et al., APS Division of Nuclear Physics Meeting, Chicago, October 2004.

^{92}Zr(d,d) versus ^{92}Zr(α, ^{3}He) -- comparison and benchmark of the surrogate method.
H. Chiang-Ai, et al., APS Division of Nuclear Physics Meeting Chicago, October 2004.

Fission yields for ^{236,238}U(d,df)
C. Plettner, et al., NUSTAR Conference, University of Surrey, U.K., January 2005.

Estimation of (n,f) cross sections by measuring reaction probability ratios
C. Plettner, H. Ai, C.W. Beausang et. al., Phys. Rev. **C 71** 051602 (2005)

Measuring reaction probability ratios to simulate neutron induced cross sections of short lived nuclei

C. Plettner, H. Ai, C.W. Beausang, et al., accepted for publication in J. Phys. G.

Other Publications

Breakdown of vibrational structure in ^{98}Ru

R.B. Cakirli, R.F. Casten, E.A. McCutchan, H. Ai, H. Amro, M. Babilon, C.W. Beausang, A. Heinz, R.O. Hughes, D.A. Meyer, C. Plettner, J.J. Ressler, N.V. Zamfir, Phys. Rev. **C70** 044312 (2004)

Transition from the seniority regime to collective motion

J.J. Ressler, R.F. Casten, N.V. Zamfir, C.W. Beausang, R.B. Cakirli, H. Ai, H. Amro, M.A. Caprio, A.A. Hecht, A. Heinz, S.D. Langdown, E.A. McCutchan, D.A. Meyer, C. Plettner, P.H. Regan, M.J.S. Sciacchitano, A.D. Yamamoto, Phys. Rev. **C69** 034317 (2004)

Isomer decay tagging in the heavy nuclei:^{210}Ra and ^{209}Ra

J.J. Ressler, C.W. Beausang, , H. Ai, H. Amro, M.A. Caprio, R.F. Casten, A.A. Hecht, S.D. Langdown, E.A. McCutchan, D.A. Meyer, P.H. Regan, M.J.S. Sciacchitano, A. Yamamoto, N.V. Zamfir, Phys. Rev. **C69** 034331 (2004)

γ*-ray spectroscopy of ^{166}Hf: X(5) in N > 90?*

E.A. McCutchan, N.V. Zamfir, R.F. Casten, M.A. Caprio, H. Ai, H. Amro, C.W. Beausang, A.A. Hecht, D.A. Meyer, J.J. Ressler, , Phys. Rev. **C 71** 024309 (2005).

Isomeric decay of ^{208}Ra

J.J. Ressler, C.W. Beausang, H. Ai, H. Amro, M. Babilon, J.A. Caggiano, R.F. Casten, G. Gurdal, A. Heinz, R.O. Hughes, E.A. McCutchan, D.A. Meyer, C. Plettner, J. Qian, M.J.S. Sciacchitano, N.J. Thomas, E. Williams, N.V. Zamfir, , Phys. Rev. **C71** 014302 (2005).

Budget

The major expenditure this year, were salary and travel to experiments and conferences