THE UNIVERSITY OF TENNESSEE

College of Arts & Sciences
Department of Physics and Astronomy
401 Nielsen Physics Building
Knoxville, TN 37996-1200
(865) 974-3342
FAX (865) 974-7843

November 22, 2013

Ms. Terri Stone
National Nuclear Security Administration
Office of Research and Development, NA-113
Room A-110, GTN
National Nuclear Security Administration
U.S. Department of Energy
1000 Independence Avenue, S.W.
Washington, DC 20585

Please find the Annual Scientific Report of the project "Theoretical Description of the Fission Process" for 2012/2013, supported by the National Nuclear Security Administration under the Stewardship Science Academic Alliances program through DOE Research Grant DE-NA0001820.

Please do not hesitate to contact me if I can provide further information.

Sincerely yours,

Witold Nazarewicz
Professor of Physics, University of Tennessee

Annual Scientific Report

"Theoretical Description of the Fission Process"
NNSA/SSAA Grant DE-NA0001820
Principal Investigator: Witold Nazarewicz (University of Tennessee)
9/14/20102-9/13/2013

1. Executive summary

Advanced theoretical methods and high-performance computers may finally unlock the secrets of nuclear fission, a fundamental nuclear decay that is of great relevance to society. Under this project, we study the phenomenon of spontaneous fission using the symmetry-unrestricted nuclear density functional theory (DFT). Our results show that many observed properties of fissioning nuclei can be explained in terms of pathways in multidimensional collective space corresponding to different geometries of fission products. From the calculated collective potential and collective mass, we estimate spontaneous fission half-lives by minimizing the collective action integral. Our calculations demonstrate that fission barriers of excited nuclei vary rapidly with particle number, pointing to the importance of shell effects even at large excitation energies. Not only does this reveal clues about the conditions for creating new elements, it also provides a wider context for understanding other types of fission.

Understanding of the fission process is crucial for many areas of science and technology. Fission governs existence of many transuranium elements, including the predicted long-lived superheavy species. In nuclear astrophysics, fission influences the formation of heavy elements on the final stages of the r-process in a very high neutron density environment. Fission applications are numerous. Improved understanding of the fission process will enable scientists to enhance the safety and reliability of the nation's nuclear stockpile and nuclear reactors. The deployment of a fleet of safe and efficient advanced reactors, which will also minimize radiotoxic waste and be proliferation-resistant, is a goal for the advanced nuclear fuel cycles program. While in the past the design, construction, and operation of reactors were supported through empirical trials, this new phase in nuclear energy production is expected to heavily rely on advanced modeling and simulation capabilities.

This was another successful year for our fission program. As documented below, a number of crucial milestones were reached and important scientific deliverables, including new science and software, were produced. Our group organized meetings on fission and extreme-scale computing and helped write white papers. These reports define several major research thrusts to be enabled by extreme scale computers that have a strong foundation in our achievements.

2. Brief summary of the goals and accomplishments of the project

The main goals of the project, can be summarized as follows:

i. Development of effective energy functionals that are appropriate for the description of heavy nuclei. Our goal is to improve the existing energy density (Skyrme) functionals to develop a force that will be used in calculations of fission dynamics. To this end, we use recently developed Hartree-Fock (HF) and Hartree-Fock-Bogoliubov (HFB) codes, see Sec. 5.C.

ii. Systematic self-consistent calculations of binding energies and fission barriers of actinide, pre-actinide, and trans-actinide nuclei using modern density functionals. This is followed by calculations of spontaneous fission lifetimes and mass and charge splits using dynamic adiabatic approaches based on the WKB approximation.

iii. Investigate novel microscopic (non-adiabatic) methods to study the fission process. In particular, we are going to assess whether the imaginary time method and the generator coordinate method can be used in practical self-consistent calculations.

iv. Develop codes and technology that can be freely used by NNSA researchers and, generally, by the low-energy nuclear physics community.

v. Train junior scientists and students to apply nuclear many-body techniques to describe low-energy nuclear phenomena.

We are pleased to report substantial progress in the all areas of the program. One measure of this progress is publications and invited material. During the reported period, our research resulted in 8 publications (Section 5.A). We presented our research in 11 invited and contributed talks at international meetings, colloquia, and seminars (Section 5.B). The results obtained under this project have been crucial for identifying microscopic description of fission as the forefront scientific problem in the era of extreme computing.

3. List of project participants

Research was carried out by the principal investigator, two research professors, two postdocs, three graduate students, and a number of collaborators. A list of personnel involved in the research covered by this grant includes:

- A. Baran, A. Staszczak and M. Warda (UTK/University of Lublin)
- J. Dobaczewski (University of Warsaw/ University of Jyväskylä)
- J. McDonnell (postdoc at LLNL; graduated from UTK in 2012)
- K. Mazurek (Institute of Nuclear Physics, Cracow, Poland)
- W. Nazarewicz (UTK/ORNL, Principal Investigator)
- E. Olsen (UTK, graduate student)
- K. Talley (UTK, graduate student)
- N. Schunck (staff at LLNL, formerly a postdoc with us)
- J. Sadhukhan (UTK, postdoc)
- J. Sheikh (UTK/ University of Kashmir Srinagar)

Four graduate students were involved in our fission research. Jordan McDonnell was a U.S. Department of Energy/NNSA Stewardship Science Graduate Fellow. His Ph.D. thesis was to describe fission pathways and level densities in highly excited nuclei. Currently, he is at LLNL as a postdoc. Erik Olsen, has been involved in developing software for our DFT and QRPA work. He was responsible for benchmarking energy density functionals in the context of proton emission and beta decay. Another graduate student involved in our fission research is Kemper Talley. Kemper is a CIRE graduate student supported by NSF, who came to us through the Bredesen Center for Interdisciplinary Research and Graduate Education. Kemper's interests in research include nuclear theory and nuclear data as it pertains to nuclear engineering problems. Currently, some of the work he is doing is concerned with updating delayed neutron codes and tables for SCALE. He is jointly supervised by Nazarewicz and Mark Williams from the Reactor Physics Group of the Reactor and Nuclear Systems Division at ORNL. Recently, we have recruited new graduate student, Noah Birge, who will join this effort.

Dr. Jhilam Sadhukhan is our new postdoc; he came from Variable Energy Cyclotron Centre, Kolkata, India. Drs. Baran, Staszczak, and Warda are senior researchers from the Maria Sklodowska-Curie University, Lublin, Poland; Dr. Dobaczewski is the head of nuclear theory group at the University of Warsaw, Warsaw, Poland and University of Jyväskylä, Finland; Dr. Katarzyna Mazurek is a young theorist from Cracow. Her background is in Langevin approach to fission. Dr. Sheikh is a Dean of the Faculty at the University of Kashmir, Srinagar, India and expert in nuclear self-consistent calculations. They all visited Tennessee for longer periods under this grant. Staszczak has been mainly responsible for our fission barrier calculations and software developments; Baran has carried out dynamic calculations of fission lifetimes and — together with Dobaczewski and Sheikh — has been working on ATDHFB framework for fission; Warda and Staszczak have collaborated with us on beta-delayed fission in the mercury-lead region. Dobaczewski is the author of the symmetry-free DFT solver HFODD used in our fission calculations.

4. Outreach

Our group has been very active, in terms of presentations, publications, and organizational involvement, in publicizing the research covered by this grant. Specifically:

- We presented the status of microscopic fission theory at several meetings, including SSAA Annual Workshop and several conferences in the U.S., Europe, Asia, and Africa (see below).
- A research topic related to the theory of fission has been listed on the website of the UTK Physics Department: http://www.phys.utk.edu/graduate/research.html - nucleartheory.
- We organized the annual fission workshops at the Joint Institute for Heavy Ion Research (see Sec. 5.D).
- We established a fission project website, http://www.phys.utk.edu/witek/fission/fission.html, that popularizes our research.
- Our work on fission barriers has been chosen as a highlight in the 2013 SSAA Annual: *"Theoretical Description of the Fission Process",* W. Nazarewicz, Stewardship Science Academic Alliances Annual 2013 DOE/NA-0019, p.11 (2013).
- Our work on optimized input for fission calculations, "Quality Input for Microscopic Fission Theory", by W. Nazarewicz, N. Schunck, and S. Wild, appeared in Stockpile Stewardship Quarterly, 2(1), 2012, p. 6.
- A 2-month program on *Quantitative Large Amplitude Shape Dynamics: Fission and Heavy Ion Fusion* by A.N. Andreyev, G.F. Bertsch, W. Loveland, and W. Nazarewicz, and W. Loveland was held at the the Institute for Nuclear Theory during September 23 - November 15, 2013, see http://www.int.washington.edu/PROGRAMS/13-3/ (see Sec. 5.E).

5. Deliverables

A. Completed projects - Publications

1. *"Fission modes of mercury isotopes",* M. Warda, A. Staszczak, and W. Nazarewicz, Phys. Rev. C **86**, 024601 (2013). Recent experiments on beta-delayed fission in the mercury-lead region and the discovery of asymmetric fission in ^{180}Hg have stimulated theoretical interest in the mechanism of fission in heavy nuclei. In this work, we studied fission modes and fusion valleys in ^{180}Hg and ^{198}Hg to reveal the role of shell effects in pre-scission region and explain the experimentally observed fragment mass asymmetry and its variation with *A*. We used the self-consistent nuclear density functional theory employing Skyrme and Gogny energy density functionals. The potential energy surfaces in multi-dimensional space of collective coordinates, including elongation, triaxiality, reflection-asymmetry, and necking, were calculated. The asymmetric fission valleys - well separated from fusion valleys associated with nearly spherical fragments - were found in in both cases considered. The density distributions at scission configurations were studied and related to the experimentally observed mass splits. We predicted a transition from asymmetric fission in ^{180}Hg towards more symmetric distribution of fission fragments in ^{198}Hg — consistent with experiment.
2. "*Spontaneous fission modes and lifetimes of superheavy elements in the nuclear density functional theory*", A. Staszczak, A. Baran, and W. Nazarewicz, Phys. Rev. C **87**, 024320 (2013). The reactions with the neutron-rich ^{48}Ca beam and actinide targets resulted in the detection of new superheavy (SH) nuclides with Z=104–118. The unambiguous identification of the new isotopes, however, still poses a problem because their α-decay chains terminate by spontaneous fission (SF) before reaching the known region of the nuclear chart. The understanding of the competition between α-decay and SF channels in SH nuclei is, therefore, of crucial importance for our ability to map the SH region and to assess its extent. In this work, we performed self-consistent calculations of the competing decay modes of even-even SH isotopes with $108 \leq Z \leq 126$ and $148 \leq N \leq 188$. We used the state-of-the-art computational framework based on self-consistent symmetry-unrestricted nuclear density functional theory capable of describing the competition between nuclear attraction and electrostatic repulsion. The collective mass tensor of the fissioning superfluid nucleus was computed by means of the cranking approximation to the adiabatic time-dependent HFB approach. Breaking axial symmetry and parity turned out to be crucial for a realistic estimate of collective action; it results in lowering SF lifetimes by more than 7 orders of magnitude in some cases. We predicted two competing SF modes: reflection symmetric modes and reflection asymmetric modes. The shortest-lived SH isotopes decay by SF; they are expected to lie in a narrow corridor formed by ^{280}Hs, ^{284}Fl, and ^{284}Uuo that separates the regions of SH nuclei synthesized in "cold-fusion" and "hot-fusion" reactions. The region of long-lived SH nuclei is expected to be centered on ^{294}Ds with a total half-life of ~1.5 days. Our survey provides a solid benchmark for the future improvements of self-consistent SF calculations in the region of SH nuclei.
3. "*Third minima in thorium and uranium isotopes in a self-consistent theory".* J. D. McDonnell, W. Nazarewicz, and J. A. Sheikh, Phys. Rev. C **87**, 054327 (2013). Well-developed third minima, corresponding to strongly elongated and reflection-asymmetric shapes associated with dimolecular configurations, have been predicted in some non-self-consistent models to impact fission pathways of thorium and uranium isotopes. These predictions have guided the

interpretation of resonances seen experimentally. On the other hand, self-consistent calculations consistently predict very shallow potential-energy surfaces in the third minimum region. In this work, we investigated the interpretation of third-minimum configurations in terms of dinuclear (cluster) states. We studied the isentropic potential-energy surfaces of selected even-even thorium and uranium isotopes at several excitation energies. In order to understand the driving effects behind the presence of third minima, we investigated the interplay between pairing and shell effects. We predicted very shallow or no third minima in the potential-energy surfaces of ^{232}Th and ^{232}U. In the lighter Th and U isotopes with N=136 and 138, the third minima seem to be better developed. We showed that the reflection-asymmetric configurations around the third minimum can be associated with dinuclear states involving the spherical doubly magic ^{132}Sn and a lighter deformed Zr or Mo fragment. We also studied isotopic chains to demonstrate the evolution of the depth of the third minimum with neutron number. We showed that the neutron shell effect that governs the existence of the dinuclear states around the third minimum is consistent with the spherical-to-deformed shape transition in the Zr and Mo isotopes around N=58. The thermal reduction of pairing, and related enhancement of shell effects, at small excitation energies help to develop deeper third minima. At large excitation energies, shell effects are washed out and third minima disappear altogether.

4. "*Microscopic Description of Nuclear Fission: Fission Barrier Heights of Even-Even Actinides*", J. McDonnell, N. Schunck, and W. Nazarewicz, in Fission and Properties of Neutron-Rich Nuclei, edited by: J H Hamilton and A V Ramayya (World Scientific, 2013), p. 597 LLNL-PROC-612272. We evaluated the performance of modern nuclear energy density functionals for predicting inner and outer fission barrier heights and energies of fission isomers of even-even actinides. For isomer energies and outer barrier heights, we find that the self-consistent theory at the HFB level is capable of providing quantitative agreement with empirical data. In particular, the recently developed UNEDF1 energy density functional yields predictions that agree well with experimental values and are on a par with, or better, than predictions of other self-consistent or macroscopic-microscopic models. While the inner barrier heights are systematically overestimated, one also needs to bear in mind that empirical values are subject to error that is at least 0.3MeV. As fit-observables are selected for future optimizations, it will be valuable to consider if additional data such as one-neutron separation energy may help to better constrain fission barrier heights.
5. "*Axially deformed solution of the Skyrme-Hartree-Fock-Bogolyubov equations using the transformed harmonic oscillator basis (II) HFBTHO v2.00d: a new version of the program,*" M.V. Stoitsov, N. Schunck, M. Kortelainen, N. Michel, H. Nam, E. Olsen, J. Sarich, and S. Wild, Comput. Phys. Commun. 184, 1592-1604 (2013). We published the new version 2.00d of the code HFBTHO that solves the nuclear HF or HFB problem by using the cylindrical transformed deformed harmonic oscillator basis. In the new version, we have implemented the following features: (i) the modified Broyden method for non-linear problems, (ii) optional breaking of reflection symmetry, (iii) calculation of axial multipole moments, (iv) finite temperature formalism for the HFB method, (v) linear constraint method based on the approximation of the Random Phase Approximation (RPA) matrix for multi-constraint calculations, (vi) blocking of quasi-particles in the Equal Filling Approximation (EFA), (vii) framework for generalized energy density with arbitrary density-dependences, and (viii) shared memory parallelism via OpenMP pragmas.
6. *"Systematic study of fission fragment mass distribution using 2-dimensional Langevin dynamics",* J. Sadhukhan and S. Pal, in Fission and Properties of Neutron-Rich Nuclei, edited

by: J H Hamilton and A V Ramayya (World Scientific, 2013), p. 564. The fragment mass distribution from fission of hot nuclei is studied in the framework of two-dimensional Langevin equations. First, the finite-range liquid drop model potential, collective inertia and one-body dissipation strength are calculated for different compound nuclei over a wide range of fissility parameter. Then, the mass asymmetry coordinate distribution is obtained from the dynamical calculation both at the saddle and the scission regions to explain the role of saddle-to-scission dynamics. Specifically, the competition between dissipative and conservative forces in determining the fragment mass distribution is investigated.

7. "*Landscape of two-proton radioactivity*", E. Olsen, M. Pfutzner, N. Birge, M. Brown, W. Nazarewicz, and A. Perhac, Phys. Rev. Lett. 110, 222501 (2013); Erratum: Phys. Rev. Lett. 111, 139903 (2013). Ground-state two-proton (2p) radioactivity is a decay mode found in isotopes of elements with even atomic numbers located beyond the two-proton drip line. So far, this exotic process has been experimentally observed in a few light- and medium-mass nuclides with $Z \leq 30$. In this study, using state-of-the-art nuclear density functional theory, we globally analyze 2p radioactivity and for the first time identify 2p-decay candidates in elements heavier than strontium. We predict a few cases where the competition between 2p emission and α-decay may be observed. In nuclei above lead, the α-decay mode is found to be dominating and no measurable candidates for the 2p radioactivity are expected.
8. *"Spontaneous fission lifetimes from the minimization of self-consistent collective action*," Jhilam Sadhukhan, K. Mazurek, A. Baran, J. Dobaczewski, W. Nazarewicz, and J.A Sheikh, Phys. Rev. C, arXiv:1310.2003. The spontaneous fission lifetime of ^{264}Fm has been studied within nuclear density functional theory by minimizing the collective action integral for fission in a two-dimensional quadrupole collective space representing elongation and triaxiality. The collective potential and inertia tensor are obtained self-consistently using the Skyrme energy density functional and density-dependent pairing interaction. The resulting spontaneous fission lifetimes are compared with the static result obtained with the minimum-energy pathway. We show that fission pathways strongly depend on assumptions underlying collective inertia. With the non-perturbative mass parameters, the dynamic fission pathway becomes strongly triaxial and it approaches the static fission valley. On the other hand, when the standard perturbative cranking inertia tensor is used, axial symmetry is restored along the path to fission; an effect that is an artifact of the approximation used. The calculation of the full ATDHFB inertia is in progress, also developments are initiated in the context of dynamical effects due to the competition between triaxial and reflection asymmetric degrees of freedom, and pairing.

B. Talks/Posters

1. "Microscopic Description of Fission Process", W. Nazarewicz, International Symposium ENSFN'12, University of Tokyo, Oct. 10-12, 2012.
2. "The nuclear landscape: theoretical perspective", W. Nazarewicz, ICFN5, Sanibel Island, FL, Nov. 4-10, 2012.
3. "Microscopic Description of Nuclear Fission: Fission Barrier Heights of Even-Even Actinides", J. McDonnell, ICFN5, Sanibel Island, FL, Nov. 4-10, 2012.
4. "Systematic study of fission fragment mass distribution using 2-dimensional Langevin dynamics", J. Sadhukhan, ICFN5, Sanibel Island, FL, Nov. 4-10, 2012.
5. "SHE: Theoretical perspective", W. Nazarewicz, Workshop focused on future studies of

super-heavy nuclei at the SHE Factory, TAMU, College Station, TX, March 12-13, 2013

6. "Low Energy Nuclear Structure and Nuclear Astrophysics", W. Nazarewicz, IUPAP WG.9 Nuclear Science Symposium, INFN - LNF, Rome, May 31, 2013
7. "Nuclear Theory and Stability: The Role of Livermorium and Flerovium", W. Nazarewicz, Celebrating Livermorium, LLNL, June 24, 2013.
8. "Study of spontaneous fission life-times using nuclear density functional theory," J. Sadhukhan/K. Mazurek, Fifth International Workshop on Nuclear fission and Fission-Product Spectroscopy, Caen, France, May 28-31, 2013.
9. "Microscopic Description of the Fission Process," J. Sadhukhan, 2013 Stewardship Science Academic Programs Annual Review Symposium, Albuquerque, NM, June 27 - 28, 2013.
10. "Density functional theory of spontaneous fission life-times", J. Sadhukhan, INT Program 13-3, on Quantitative Large Amplitude Shape Dynamics: fission and heavy ion fusion, Oct. 2, 2013.
11. "Stability and properties of heavy and superheavy nuclei in mean-field model with Skyrme energy density functional", A. Staszczak, INT Program 13-3, on Quantitative Large Amplitude Shape Dynamics: fission and heavy ion fusion, Oct. 17, 2013.

C. Code developments

As stated in our original proposal, "*Developing codes and technology that can be freely used by NNSA is also one of our goals*." We developed an accurate 2D lattice Skyrme-HFB solver HFBAX based on B-splines. We used this code to study fission barriers. In addition to providing new physics insights, HFBAX can serve as a useful tool to assess the reliability and applicability of coordinate-space and configuration-space HFB solvers, both existing and in development. Another code developed by our group is HFODD (v2.49t), which solves the Skyrme-Hartree-Fock-Bogoliubov equations in the Cartesian-deformed harmonic-oscillator basis. Version 2.49t of HFODD provides a number of new options such as the isospin mixing and projection of the Skyrme functional, the finite-temperature HF and HFB formalism and optimized methods to perform multi-constrained calculations. It is also the first version of HFODD to contain threading and parallel capabilities.This code is currently used in all our advanced fission calculations that require breaking of most self-consistent symmetries. Based on HFODD, we developed codes to calculate collective inertia (tensor of mass parameters). Finally, we published an axial HFB solver HFBTHO, which uses the transformed harmonic oscillator basis. In the new version 2.00d of the code, we developed shared memory parallelism via OpenMP pragmas.

D. **Annual workshop.** As stated in our original proposal, "*We will hold workshops at the Joint Institute for Heavy Ion Research devoted to the fission problems that we will pursue under this proposal. We will solicit input from NNSA laboratory researchers on what is relevant to calculate*." In 2007, 2008, 2009, 2010, 2011, and 2012 we held Joint JUSTIPEN-LACM Meetings at the Joint Institute for Heavy Ion Research in Oak Ridge. The meeting in 2007 was a merger of two workshops: (i) the US-Japan theory meeting under the auspices of the Japan-US Theory Institute for Physics with Exotic Nuclei (JUSTIPEN), and (ii) the annual NNSA-JIHIR meeting on the nuclear large-amplitude collective motion (LACM) with an emphasis on fission. The workshops were very well attended (60 participants in 2007, over 70 participants in 2008, 90 participants in 2009, 60 participants in 2010, 62 participants in 2011, and 60 participants in 2012) and involved participants from NNSA/DP Laboratories (LANL, LLNL, NNSA), as well as students and post-docs. The workshops were partly sponsored by this grant, and reference to NNSA support was displayed during the meeting.

E. **INT 13-3 Program on fission**. The INT program on Quantitative Large Amplitude Shape Dynamics: fission and heavy ion fusion was held in Seattle, WA from 23 September to 15 November, 2013. The organizers were: A.N. Andreyev (University of York), G.F. Bertsch (Institute for Nuclear Theory), W. Loveland (Oregon State University), and W. Nazarewicz (University of Tennessee/ORNL). The intent of this program was to: (i) bring together theorists working on predictive theories of the underlying shape dynamics to compare various approaches and computational methodologies; (ii) actively foster collaborations between the major actors in the field, including collaborations between experiment and theory; (iii) identify critical experimental data that will inform theoretical developments; and (iv) identify strategies and resources to break computational barriers in this area. We believe that excellent progress has been made in all four areas. The program was well attended, with 65 total participants. Week 4 of the program was organized as predominantly experimental workshop, which dealt largely with new data on fission and fusion to challenge nuclear theory. The talks at INT-13-3 can be viewed here: http://www.int.washington.edu/talks/WorkShops/int_13_3/. The program needs to be put into the perspective of the intellectual development that occurred since the last INT program on large-amplitude dynamics, INT-95-3 (organized by Bertsch and Nazarewicz). As presented in the talks, we now have accurate descriptions (< 1 MeV) of fission barriers based on self-consistent mean-field theory, with the beginnings of systematic error assessments of the theory. Quantitative results are now available the calculation of heavy–ion fusion cross sections by TDHF. The calculations of spontaneous fission lifetimes have become much more sophisticated in the context of the WKB barrier penetration model: more collective degrees of freedom are taken into account in setting the fission path; the path may now be determined by minimizing the action; the inertia term in the action integral may be treated in a way consistent with the instanton formulation of barrier penetration. We also saw a major advance in the theory of fission mass distributions: many of the details can be reproduced by a statistical treatment with diffusive dynamics. We expect that new experimental efforts discussed during the program will not only greatly increase the database for which current theory can be applied but will also provide its crucial tests. An initial goal of the program, to reevaluate the basic assumption in the theory of nuclear fission, is being realized by a position paper being drafted to investigate a new approach to making a theory that is both broad enough to treat the adiabatic and the dissipative limits of fission dynamics, as well as being computationally tractable. A reporting guide is being prepared for theorists to make it easier to compare different methods and to assess the reliability of theory for applications. The guide includes the list of measured/evaluated quantities (spontaneous fission half-lives, fission barrier heights, fission fragment mass distributions, and excitation energies of fission isomers) that can be used to benchmark various methodologies and optimize models. The guide also gives recommendations on error analysis of the theory and how to present the uncertainties.