LLNL-PROC-400347

LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Microscopic Theory of Fission

W. Younes, D. Gogny

January 9, 2008

Compound Nuclear Reactions and Related Topics
Fish Camp, CA, United States
October 22, 2007 through October 26, 2007

Microscopic Theory of Fission

W. Younes and D. Gogny

Lawrence Livermore National Laboratory, Livermore, CA 94550

Abstract. In recent years, the microscopic method has been applied to the notoriously difficult problem of nuclear fission with unprecedented success. In this paper, we discuss some of the achievements and promise of the microscopic method, as embodied in the Hartree-Fock method using the Gogny finite-range effective interaction, and beyond-mean-field extensions to the theory. The nascent program to describe induced fission observables using this approach at the Lawrence Livermore National Laboratory is presented.

Keywords: fission,Hartree Fock Bogoliubov,Gogny interaction,fission fragments,Plutonium
PACS: 21.60.Jz,25.85.-w,25.85.Ec,27.90.+b

INTRODUCTION

The quantitative description of fission is arguably the most daunting challenge in nuclear physics. Since the official discovery of fission in 1939 [1], a predictive theory of this phenomenon has remained elusive. However, recent developments in the formalism, coupled with the advent of parallel programming, have made the microscopic treatment of fission within the framework of quantum many-body theory feasible. The microscopic approach to fission, embodied in the Hartree-Fock (HF) method, and augmented by a set of extensions beyond the mean-field approximation (e.g., the Bogoliubov formalism used to include pairing correlations, the random-phase approximation used to include residual particle-hole correlations, etc.), is the only formalism that is potentially capable of producing the long-sought-after predictive theory of this phenomenon.

The tremendous challenge posed by the development of a quantitative, predictive theory of fission stems from a variety of underlying difficulties. In particular: 1) *Fission is a true quantum many-body problem*. The quantitative description of the fission process, and the resulting observables depend very sensitively on the microscopic details. 2) *Fission is a dynamic phenomenon*. Descriptions of fission (e.g., based on the liquid-drop model of the nucleus with quantum shell corrections) that are based solely on a potential energy surface calculation ignore the dynamical response of the nucleus (i.e., the inertial tensor), which varies as its configuration changes. 3) *Fission involves large-amplitude collective motion*. From formation to scission, the nucleus explores a variety of exotic shapes, ultimately driving the single system into two (or more) separate fragments. Thus, a unified approach is needed to describe in a consistent manner the full gamut of relevant configurations. Collective variables play a critical role in providing this unified description. 4) *Fission involves both single-particle and collective degrees of freedom, and their coupling*. For low-energy induced fission (e.g., for a nucleus formed up to about 2 MeV above the first barrier) There is insufficient energy to significantly excite intrinsic states of the nucleus. Therefore the motion remains adiabatic, for the most part, as the nucleus evolves toward scission. For higher-energy fission, however, this is no longer a good approximation.

On the other hand, the payoff from meeting the challenge posed by the theoretical description of fission is correspondingly high. The ability to predict fission observables reliably where they cannot be measured is of immediate importance to applied-physics efforts, be they in industry, nuclear energy, or Homeland security. Furthermore, the theoretical understanding of fission implies the understanding of a variety of fundamental nuclear-physics phenomena. The same theoretical framework used to describe fission can be used as a starting point for the study of some aspects of fusion reactions, adiabatic versus non-adiabatic phenomena (coupling between intrinsic and collective degrees of freedom), the transition from single-particle to collective degrees of freedom, shape coexistence, shape isomerism, etc.

THE MICROSCOPIC APPROACH TO FISSION

The outcome of the fission process depends sensitively on the microscopic details of the problems. As an extreme example, we note that the measured lifetimes for spontaneous fission of ^{256}Fm and ^{258}Fm differ by seven orders of magnitude. Because the details of the energy surface can change significantly from one nucleus to the next, a predictive theory of fission must be constructed from building blocks that are predetermined for all nuclei. This is the basic principle behind the microscopic approach.

In the microscopic approach, the fissioning nucleus is built up only from its constituent neutrons and protons and an effective (i.e., in-medium) interaction between them. For tractability, this many-body problem is solved in the Hartree-Fock-Bogoliubov approximation. The Hartree-Fock (HF) method provides an approximate solution to the many-body problem with two important features: i) for many applications, it constitutes a very good first approximation, and ii) there are well-established, rigorous procedures for improving the HF solution and restoring the physics missing in the approximation. In particular, the Hartree-Fock-Bogoliubov (HFB) formalism goes beyond the mean-field approximation of HF to include residual pairing interactions between nucleons.

The input to the HF procedure is the effective interaction between the nucleons, used to generate the mean field. The mathematical form of this interaction is constrained, but not completely and uniquely defined by symmetry requirements (e.g., rotational and translational invariance). Therefore an explicit form must be postulated for this interaction, and its parameters must be fixed by a fit to experimental observables. The general form for the effective interaction consists of a central part, a spin-orbit interaction, a density-dependent contribution, and a Coulomb interaction which is applied only between protons. The density-dependent part of the force is motivated by more fundamental approaches, such as the Brueckner G-matrix theory, which describes the nucleon-nucleon interaction in the presence of other nucleons, using an approach that is formally similar to that of scattering theory. The work described in this paper uses the finite-range interaction developed by D. Gogny [2]. The use of a finite-range interaction is of paramount importance, as it allows the mean field and pairing field to be treated on the same footing. In other words, the same interaction that gives rise to the mean field is used to generate the residual pairing interaction between the nucleons, in a completely self-consistent manner. Pairing plays a critical role in the description of heavy nuclei and fission, and the ability to include it in a natural way is crucial.

The HFB procedure can be extended to describe collective motion of the nucleus. Small-amplitude motions (e.g., low-energy vibrations of the nuclear surface) give rise to particle-hole excitations across the Fermi surface, and the resulting residual interactions between particles and holes (as well as those particle-particle and hole-hole correlations beyond pairing) can be taken into account by the Quasiparticle Random-Phase Approximation (QRPA). For large-amplitude collective motion that occur in fission, the nucleus can explore shapes far from that given by the HFB solution. The HFB method is designed to find a single particular shape of the nucleus, the one that minimizes its energy, but can readily be extended by introducing external fields to yield solutions for any desired nuclear shape. The Constrained Hartree-Fock-Bogoliubov (CHFB) method introduces the required external fields via Lagrange multipliers. Each CHFB solution is a single Slater determinant for a given nuclear shape, but a large-amplitude collective motion of the nucleus consists of a mixture of all these solutions. The Generator-Coordinate Method (GCM) constructs such a linear superposition of the CHFB solutions through a variational procedure. The Time-Dependent Generator-Coordinate Method (TDGCM) is a further extension that constructs a wave packet from the CHFB solutions and describes it time evolution to scission, giving a fully quantum-mechanical , time-dependent description of fission. When TDGCM calculations are combined with QRPA corrections on the HFB states, dynamic calculations of fission can be performed with an unprecedented level of sophistication that includes crucial correlations in the HFB solutions.

The TDGCM also provides a formalism within which intrinsic excitations of the nucleus (e.g., two-quasiparticle, four-quasiparticle, etc.) can be treated. These excitations and their couplings are needed to describe fission at intermediate energies (e.g., for equivalent incident neutron energies of up to $\sim$ 10 MeV). At higher energies still, a temperature-dependent formalism becomes more appropriate, but with increasing temperature the fission process is expected to eventually transition to something more like fragmentation, where the nucleus fractures without elongating, and the collective degrees of freedom are expected to play a diminishing role in the description of the fission process.

PAST ACHIEVEMENTS

In a series of articles by Berger *et al.* [3, 4, 5], the microscopic method was used to explain some fundamental aspects of the fission process, by demonstrating the importance of collective degrees of freedom in fission. The authors studied the fission of ^{240}Pu using two collective variables; the quadrupole moment Q_{20} (related to the elongation of the nucleus), and the hexadecapole moment Q_{40} of the nucleus (related to the thickness of the neck separating the nascent fragments). In their calculations, the octupole moment Q_{30}, related to the mass asymmetry of the fission fragments, was not constrained, and therefore assumed the value for the most likely split (i.e., around ^{106}Mo/^{134}Te).

Berger *et al.* discovered that over a wide range of elongations of the nucleus, from $Q_{20} \approx 250$ b to 400 b, two HFB minima coexisted, but with very different values of Q_{40}. The first set of solutions, with larger values of Q_{40} and higher energies, corresponds to configurations of the nucleus that stretches along its symmetry axis with increasing Q_{20} without breaking, and is dubbed the "fission valley". The second set of solutions, with smaller values of Q_{40} and lower energies, corresponds to a nucleus that is always broken into a pair of well-separated fragments, and forms the "fusion valley". In the $Q_{20} - Q_{40}$ plane, these two valleys are separated by a barrier that is never higher than ~ 5 MeV, and disappears with increasing Q_{20}. Near $Q_{20} \approx 400$ b, the barrier separating the two solutions disappears entirely, and the nucleus spontaneously drops into the fusion valley, dividing into two fragments.

The scission of the nucleus at high elongation, where the barrier between the fission and fusion valleys disappears corresponds to phenomenon known as "hot" fission. In this situation, the fragments are formed relatively far apart due to the ~ 400 b elongation of the parent nucleus, and their mutual kinetic energy is consequently smaller, leading in turn to the formation of comparatively highly excited, hot fragments. Conversely, it is possible for the nucleus to overcome the barrier between the two valleys at lower elongations of the nucleus. In an extreme case, the fragments are formed closer together, their relative kinetic energy is higher, and their respective excitation energies are essentially zero. This is the "cold-fission" phenomenon observed experimentally [6, 7]. In between the hot and cold exit points, the same fragments can be formed with a plethora of possible excitation and kinetic energies. Thus, the addition of collective variables in the microscopic calculation leads naturally to the prediction of a path dependence of the fission process.

The identification of the mechanism that allows the nucleus to overcome the barrier in the $Q_{20} - Q_{40}$ plane separating the fission and fusion valleys, is itself another triumph of the microscopic theory of fission, which lead to the quantitative prediction of reasonable fission times. Contrary to expectations, tunneling is not the process by which the nucleus transits from the fission to the fusion valley. Rather, it is the coupling between collective variables that is responsible for the transition. Schematically, as the nucleus moves along the fission valley, the coupling between the Q_{20} and Q_{40} degrees of freedom allows energy to be exchanged between the two. As a result, the longitudinal motion of the nucleus along the fission valley slows down, while the transverse motion speeds up, "pumping up" the nucleus through collective excitations in the local potential well of the fission valley. As the nucleus is excited ever higher by the $Q_{20} - Q_{40}$ coupling, it eventually rises above the fission-fusion barrier, and drops into the lower fusion valley. This schematic description is borne out by rigorous TDGCM calculations performed for ^{240}Pu [3, 4, 5], that produced comparatively-long characteristic scission times of $\sim 3 \times 10^{-21}$ s. Previously, unacceptably short scission times had been the bane of non-microscopic descriptions of fission, leading to the introduction of ad-hoc "dissipation" terms to slow down the scissioning nucleus. By contrast, the microscopic description of fission produces these longer times naturally, without the need for any extraneous mechanisms.

More recently [8], HFB + TDGCM calculations have been performed to describe the low-energy fission of ^{238}U. The fragment mass yields and kinetic energies in particular were found to be in remarkable agreement with experimental data. In the most recent work by the BIII group [9], the technique has been applied to calculate fission-fragment properties for ^{226}Th and ^{256,258,260}Fm.

THE LLNL MICROSCOPIC FISSION PROGRAM

An ambitious program has been started at LLNL to develop a microscopic picture of induced fission with a particular emphasis of the fission-fragment properties, based on the highly successful work at BIII. This work is a "spiritual kin" to the ongoing program at BIII, discussed elsewhere in these proceedings by N. Dubray.

In early 2007, the HF code FRANCHFRI [10] (Finite-RANge Constrained Hartree-Fock Rapid Iterator) was completed, and served as the starting point to develop the HFB code FRANCHBRIE [11] (Finite-RANge Constrained Hartree-Fock-Bogoliubov with Rapid Iteration Execution). The code FRANCHBRIE is the main tool that will be used in static fission calculations at LLNL. The code calculates two-body matrix elements of the Gogny interaction using the very efficient separation method developed by D. Gogny [12]. The separation method expresses the two-body matrix elements as a finite sum of products of one-body matrix elements. The code assumes axial and time-reversal symmetries, but reflection symmetry can be broken. Matrix elements are calculated in a one-center harmonic-oscillator basis, but following the prescription of Warda *et al.* [13], different number of oscillator shells are allowed in the radial and z directions. In this way, it is possible to describe nuclear shapes highly elongated along the z axis, using manageable basis sizes. The code also uses expressions derived by Egido et al. [14] for improved numerical stability in the application of the separation method. The HFB calculations can be constrained to give specific average numbers of protons and neutrons, the moments $Q_{\mu 0}$ for $\mu = 1-4$, the separation between the fragments, as well as the average number of protons and neutrons for the left and right fragments separately. The code also offers a certain number of options that trade off execution speed for accuracy: the Coulomb exchange contribution can be calculated exactly or in the Slater approximation, the Coulomb interaction can be included

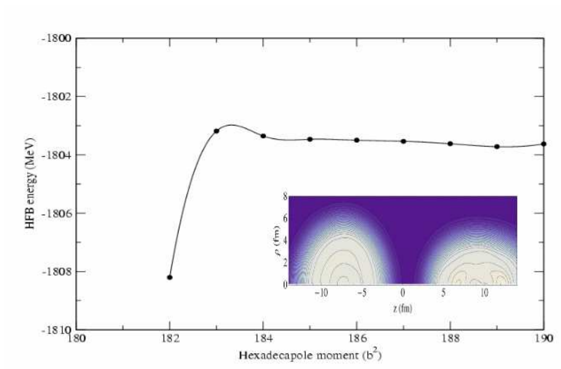

Figure 1. HFB energy calculated using FRANCHBRIE for ^{240}Pu, as a function of Q_{40}, for Q_{20} constrained to 340 b, and Q_{30} constrained to 46 b$^{3/2}$. The inset shows a contour plot of the nuclear density for $Q_{40} = 182\,b^2$, the first point on the plot for which the neck connecting the two fragments disappears.

in the pairing-field calculation or not, and the two-body center-of-mass correction term can be included in the mean-field calculation or not. A parallel version of the code, FRANCHBRIE$_{\parallel}$, has also been implemented to produce large-scale HFB calculations on a grid of constraint values.

The code is currently being used to identify scission configurations and the associated fission-fragment properties (e.g., kinetic and excitation energies, shapes of the fragments). Fig. 1 shows an HFB calculation at scission. With Q_{20} and Q_{30} fixed, Q_{40} is decreased in steps of $1\,b^2$ until the neck separating the nascent fragments disappears; this happens rather suddenly at $Q_{40} = 182\,b^2$. At that point, the densities of the fragments can be integrated giving particle numbers consistent with a ^{135}I/^{105}Nb mass split, and a separation of 16.9 fm between the fragments. The Coulomb energy between the fragments, which can be equated with their relative kinetic energy is therefore 185.5 MeV. The energy densities can be integrated for each fragment, and compared to HFB calculations of their ground-state energies, to give excitation energies of 10.2 and 8.7 MeV for the ^{135}I and ^{105}Nb fragment, respectively. As a rough estimate of neutron multiplicity, dividing these excitation energies by the average energy lost in neutron evaporation gives $\bar{\nu} = 1.04$ for ^{135}I and 1.04 for ^{105}Nb. These preliminary calculations will be repeated for many other configurations, in order to form a more complete picture of the properties of fission fragments at scission.

CONCLUSION

The microscopic method is, at present, the most promising framework for producing a predictive theory of fission. In the microscopic approach, embodied in the Hartree-Fock formalism and its extensions, the only phenomenological input to the theory is the effective interaction between nucleons. The parameters of this interaction are fitted to a handful of nuclear data, and validated through quantitative predictions of a wealth of observables throughout the nuclear chart. The approximations used in the microscopic formalism are well-understood and can be lifted in a rigorous manner, albeit at some computational cost. In recent years, the microscopic approach has achieved notable success in the description of fission, such as the prediction of cold and hot fission modes, and realistic fission-time calculations. A program is currently underway at LLNL to leverage the ongoing success of the microscopic theory, and implement a systematic study of induced fission, and of the associated properties of fission fragments.

ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344.

REFERENCES

1. O. Hahn and F. Strassmann, Naturwiss. **27**, 11-15 (1939).
2. D. Gogny, Proceedings of the International Conference on Nuclear Selfconsistent Fields, Trieste, 333-352 (1975).
3. J. F. Berger, M. Girod, and D. Gogny, Nucl. Phys. **A428**, 23-36 (1984).
4. J. F. Berger, M. Girod, and D. Gogny, Nucl. Phys. **A502**, 85-104 (1989).
5. J. F. Berger, M. Girod, and D. Gogny, Comp. Phys. Comm. **63**, 365-374 (1991).
6. C. Signarbieux, M. Montoya, M. Ribrag, C. Mazur, C. Guet, P. Perrin, and N. M. Maurel, J. Phys. Lett. **42**, 437-440 (1981).
7. M. Ashgar, N. Boucheneb, G. Medkour, P. Geltenbort, and B. Leroux, Nucl. Phys. **A560**, 677-688 (1993).
8. H. Goutte, J. F. Berger, P. Casoli, and D. Gogny, Phys. Rev. C **71**, 024316 (2005).
9. N. Dubray, H. Goutte, and J.-P. Delaroche, accepted for publication in Phys. Rev. C (2007).
10. W. Younes and D. Gogny, LLNL Tech. Rep. UCRL-TR-227645 (2007).
11. W. Younes and D. Gogny, LLNL Tech. Rep. UCRL-TR-234682 (2007).
12. D. Gogny, Nucl. Phys. **A237**, 399-418 (1975).
13. M. Warda, J. L. Egido, L. M. Robledo, and K. Pomorski, Phys. Rev. C 66, 014310 (2002).
14. J. L. Egido, L. M. Robledo, and R. R. Chasman, Phys. Lett. B 393, 13-18 (1997).