

Uncertainty Quantification for Nuclear Density Functional Theory and Information Content of New Measurements

J. D. McDonnell,^{1,2} N. Schunck,² D. Higdon,³ J. Sarich,⁴ S. M. Wild,⁴ and W. Nazarewicz^{5,6,7}

¹*Department of Physics and Astronomy, Francis Marion University, Florence, South Carolina 29501, USA*

²*Physics Division, Lawrence Livermore National Laboratory, Livermore, California 94551, USA*

³*Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*

⁴*Mathematics and Computer Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA*

⁵*Department of Physics and Astronomy and NSCL/FRIB Laboratory, Michigan State University, East Lansing, Michigan 48824, USA*

⁶*Physics Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA*

⁷*Institute of Theoretical Physics, Faculty of Physics, University of Warsaw, ul. Hoża 69, 00-681 Warsaw, Poland*

(Received 15 January 2015; published 24 March 2015)

Statistical tools of uncertainty quantification can be used to assess the information content of measured observables with respect to present-day theoretical models, to estimate model errors and thereby improve predictive capability, to extrapolate beyond the regions reached by experiment, and to provide meaningful input to applications and planned measurements. To showcase new opportunities offered by such tools, we make a rigorous analysis of theoretical statistical uncertainties in nuclear density functional theory using Bayesian inference methods. By considering the recent mass measurements from the Canadian Penning Trap at Argonne National Laboratory, we demonstrate how the Bayesian analysis and a direct least-squares optimization, combined with high-performance computing, can be used to assess the information content of the new data with respect to a model based on the Skyrme energy density functional approach. Employing the posterior probability distribution computed with a Gaussian process emulator, we apply the Bayesian framework to propagate theoretical statistical uncertainties in predictions of nuclear masses, two-neutron dripline, and fission barriers. Overall, we find that the new mass measurements do not impose a constraint that is strong enough to lead to significant changes in the model parameters. The example discussed in this study sets the stage for quantifying and maximizing the impact of new measurements with respect to current modeling and guiding future experimental efforts, thus enhancing the experiment-theory cycle in the scientific method.

DOI: [10.1103/PhysRevLett.114.122501](https://doi.org/10.1103/PhysRevLett.114.122501)

PACS numbers: 21.10.Dr, 21.60.Jz, 24.75.+i, 02.30.Zz

Introduction.—Our understanding of heavy, complex nuclei lies at the heart of many basic science questions, such as chemical evolution, neutron star structure, synthesis of superheavy elements, mechanism of nuclear fission, or search for the new standard model [1]; this knowledge is also crucial for societal applications [2]. In all those cases, reliable theoretical estimates of nuclear masses, low-lying excitations, electromagnetic strength, and nuclear reaction rates form essential inputs when direct experimental information is not available.

For tackling complex nuclei theoretically, nuclear density functional theory (DFT) is the microscopic tool of choice [3]. In recent years, largely because of algorithmic developments and high-performance computing [4], DFT has taken great strides as a predictive theory that describes the properties of nuclei across the nuclear landscape [5–7]. No consensus exists, however, on the form of the nuclear effective interaction or energy density functional (EDF), resulting in large systematic uncertainties. Moreover, nuclear EDFs are characterized by coupling constants that must be adjusted to experiment [3,8–10]. The systematic calculation of uncertainties related to the determination of

model parameters, as well as the propagation of these uncertainties in model prediction, has thus become a necessity [9,11–15] (see also Ref. [16]). Furthermore, as we enter the era of experiments with exotic nuclei at extremes of isospin, theory will play an increasingly important role in identifying scientific priorities of planned experimental campaigns. Conversely, as experiments extend current knowledge by providing information about the uncharted regions of the nuclear landscape, new methodologies become critical for evaluating the impact of these measurements on theory.

From the viewpoint of statistics, determining the parameters of a model given a set of experimental data measurements is an inverse problem [17]. Bayesian inference methods [18] are one of the most popular and powerful statistical approaches to inverse problems, with diverse applications in physics [19,20] (for recent nuclear physics applications, see, e.g., Refs. [21–27]). In the Bayesian setting, model parameters are treated as random variables, and their uncertainty is characterized by their joint probability distribution. Various techniques, often based on Monte Carlo simulations, have been developed to

reconstruct this probability distribution from model prediction of experimental data.

Objectives.—In this work, we present the advanced application of Bayesian inference to global nuclear properties using nuclear DFT. In particular, we use the Bayesian framework to quantify and propagate DFT statistical model uncertainties and to assess the information content of new data with respect to model developments. To this end, we study the impact of the recently reported mass measurements from the Canadian Penning Trap (CPT) mass spectrometer at Argonne National Laboratory [28–30] on the Bayesian posterior probability distribution as well as the direct determination of EDF parameters. The CPT data set is unique in that it probes neutron-rich nuclei around ^{132}Sn ; hence, it can help improve our knowledge of isovector EDF properties and reduce extrapolation uncertainties into the region of the astrophysical r process. From the resulting posterior distribution, we assess model uncertainties on observables, including the position of the two-neutron dripline and fission barrier heights of actinide nuclei.

Method.—Our theoretical framework is nuclear density functional theory with Skyrme EDFs. Pairing is modeled with a density-dependent pairing force and treated at the Hartree-Fock-Bogoliubov (HFB) level by using an approximate particle number projection with the Lipkin-Nogami method. We choose the UNEDF1 parametrization of the Skyrme functional as our reference model [31]. This EDF is characterized by twelve parameters that were optimized on a set of binding energies for spherical and deformed nuclei, charge radii, odd-even mass differences, and excitation energies of selected fission isomers (see Refs. [31–33] for details of the model and the UNEDF EDF family).

The quality of the functional is measured by a composite χ^2 function,

$$\chi^2(\mathbf{x}) = \frac{1}{n_d - n_x} \sum_{t=1}^{n_T} \sum_{j=1}^{n_t} \left(\frac{y_{tj}(\mathbf{x}) - d_{tj}}{\sigma_t} \right)^2, \quad (1)$$

where \mathbf{x} denotes the set of model parameters, $n_x = 12$ is the number of model parameters, n_T the number of different data types used in the fit ($n_T = 4$ in our case), n_t is the number of data points used for each data type, $n_d = \sum_t n_t$ is the total number of data points, and d_{tj} and $y_{tj}(\mathbf{x})$ are the experimental value and corresponding model prediction, respectively, for the j th data point of type t . For the UNEDF1 functional, where $n_d = 115$, computing the χ^2 requires about 5 min of CPU time with over 800 cores in a multithreaded implementation of the DFT solver HFBTHO [34]. Monte Carlo simulations used to construct the posterior distribution may typically involve tens of thousands of such χ^2 evaluations; even with current supercomputers, this cost is too high. We thus replace the DFT model $y_{tj}(\mathbf{x})$ with a Gaussian process (GP)

response surface, allowing Monte Carlo based Bayesian computation.

The GP response surface is estimated within the encompassing Bayesian formulation [35] by using an ensemble of “training” DFT computations for each of the n_d experimental nuclei used in (1). We constructed our training ensemble with 200 DFT computations, each with a randomly sampled set of n_x Skyrme parameters. The $200 \times n_x$ Skyrme parameters were distributed over an n_x -dimensional hyper rectangle centered on the UNEDF1 values following the algorithm given in [36]. For each parameter, widths are determined according to the standard deviations reported in Ref. [31], which were obtained through a covariance analysis that assumed a linear approximation. Thus the parameter ensemble is distributed according to the prior distribution for the Skyrme parameters—uniform over this n_x - d hyper rectangle. The GP is controlled by a scaling parameter, as well as correlation parameters regulating the smoothness of the response surface in each of the n_x parameter directions.

From the GP response surface we calculate the posterior distribution, which provides the probability that a given model parameter set will reproduce the known experimental data. The full posterior density includes a likelihood term for the experimental data based on Eq. (1) and the ensemble of training DFT computations for the GP, the uniform prior for the model parameters \mathbf{x} , and priors for the parameters that control the GP-based response surface; see Ref. [37] for a detailed description of the posterior density. We construct dependent samples from this distribution using Markov chain Monte Carlo calculations as detailed in [35], from which summaries such as 90% probability intervals and posterior means can be constructed.

Results.—Through Bayesian model calibration, we first obtained the posterior probability distribution for the UNEDF1 parameter set, which provides a sense of how the set of fit observables of UNEDF1 constrains the parameters. In Fig. 1, we show the univariate and bivariate marginal estimates of the posterior distribution. The blue-outlined regions give the 95% posterior probability region for the original UNEDF1 parameters. We notice that the Bayesian approach is in agreement with estimates of uncertainties based on covariance analysis reported in Ref. [31]. In particular, most distributions are centered on the UNEDF1 values, and the standard deviations extracted from the distribution are consistent with the 95% probability intervals.

In a second step, we used our Bayesian formulation to evaluate the information content of the new mass measurements [28–30]. To this end, we modified the χ^2 of Eq. (1) to include 17 new masses of neutron-rich even-even nuclei measured at the CPT; the experimental values are listed in the Supplemental Material [38]. The GP response surface was again produced by using an augmented

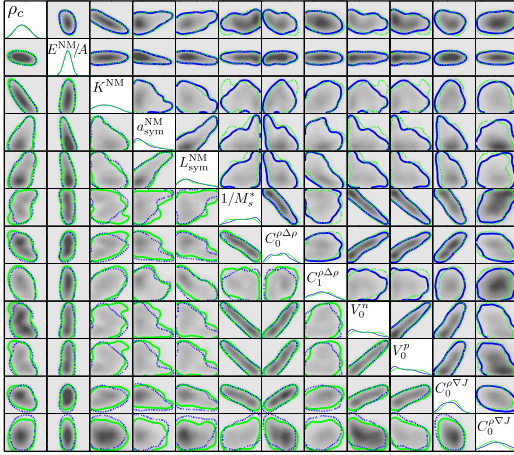


FIG. 1 (color online). Univariate and bivariate marginal estimates of the posterior distribution for the 12-dimensional DFT parameter vector of the UNEDF1 parametrization. The blue lines enclose an estimated 95% region for the posterior distribution found when only the original UNEDF1 data are accounted for; the green-outlined regions represent the same region for the posterior distribution found when the new CPT mass measurements are included. The ranges of parameter variations are $0.155 \leq \rho_c \leq 0.165$ (fm^{-3}); $-16.0 \leq E^{NM}/A \leq -15.5$ (MeV); $200 \leq K^{NM} \leq 240$ (MeV); $28 \leq a_{\text{sym}}^{NM} \leq 30$ (MeV); $20 \leq L_{\text{sym}}^{NM} \leq 60$ (MeV); $0.8 \leq 1/M_s^* \leq 1.2$; $-60 \leq C_0^{\rho\Delta\rho} \leq -40$ (MeV fm^5); $-200 \leq C_1^{\rho\Delta\rho} \leq -90$ (MeV fm^5); $-200 \leq V_0^n \leq -150$ (MeV fm^3); $-220 \leq V_0^p \leq -180$ (MeV fm^3); $-80 \leq C_0^{\rho\nabla J} \leq -60$ (MeV fm^5); and $-80 \leq C_1^{\rho\nabla J} \leq 0$ (MeV fm^5).

ensemble of $200(n_d + 17)$ DFT model evaluations. The green-outlined regions in Fig. 1 represent the same 95% posterior probability regions obtained with the inclusion of the Argonne mass measurements. With the exception of a few ill-constrained parameters (e.g., nuclear incompressibility and isovector surface coupling constant), the shift in the posterior is small for each parameter. This suggests a weak impact of the additional data on our model.

For comparison, we performed a direct reoptimization, independent of the GP response surface, of the UNEDF1 functional that includes the new CPT masses [39]. We refer to the reoptimized EDF parameter set as UNEDF1_{CPT}; see Supplemental Material for parameter values [38]. The two parametrizations are similar. The largest relative difference, weighted by the standard deviations reported in Ref. [31], is 0.6σ for the isovector surface coupling constant $C_1^{\rho\Delta\rho}$ and surface symmetry energy. These quantities have been difficult to constrain with the data set used in the UNEDF protocol. Of interest, then, is the fact that the UNEDF1_{CPT} value ($C_1^{\rho\Delta\rho} = -114.2915$) is close to that of UNEDF2 ($C_1^{\rho\Delta\rho} = -113.164$), which was also optimized to effective single-particle energies [33], known to be sensitive probes of surface properties. Since the new data set including CPT masses is more skewed toward neutron-rich nuclei, it may supply additional information about the shell structure

TABLE I. Root-mean-square deviations for each of the types of data included in the UNEDF optimization. Masses and energies are in MeV, radii in fm.

Class	UNEDF1	UNEDF1 _{CPT}
Masses (def)	0.721	0.578
Masses (sph)	1.461	1.545
Radii	0.022	0.022
Odd-even staggering (n)	0.023	0.024
Odd-even staggering (p)	0.079	0.081
Fission isomer energies	0.190	0.316
Masses (CPT)	1.064	0.479

above doubly magic ^{132}Sn through a better determination of isovector coupling constants.

Table I displays the root-mean-square deviation between calculated and measured values for each type of data included in the optimization. We note that the inclusion of the CPT mass measurements shifts the optimization priority, so that the new masses and deformed masses are reproduced more accurately, while predictions for fission isomers and spherical masses deteriorate slightly. The results in Table I are indicative of a small, additional constraint on the isovector coupling constants in UNEDF1_{CPT}.

Equipped with the posterior distribution for the EDF parameters, we now turn to the propagation of statistical uncertainties for model predictions. Implicit in the χ^2 of Eq. (1) is the statistical model for a new mass measurement $d_{ij} = y_{ij}(\mathbf{x}) + \varepsilon_{ij}$, with the “model misfit” error term ε_{ij} assumed to be independent, following a normal distribution $N(0, s_i^2)$. We take the posterior distribution for the EDF parameters obtained by conditioning only on the measurements used in the fit of UNEDF1 and propagate the distribution through the augmented GP-based emulator,

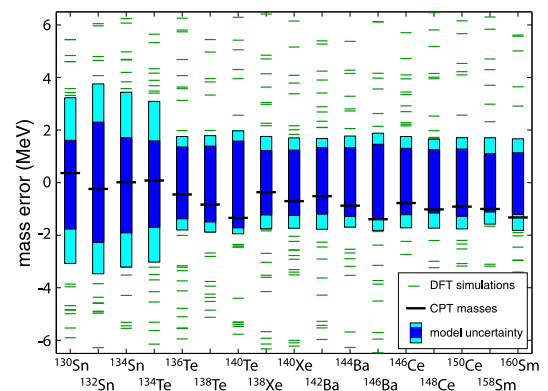


FIG. 2 (color online). Estimated theoretical error bars for the masses of the even-even nuclei measured in Refs. [28–30], using the posterior distribution for UNEDF1. Thin green bars show the results of training DFT calculations. Dark blue bands represent the 90% confidence bands due to uncertainty in the EDF parameters; larger, light blue bands also account for model error; black bars show mass residuals.

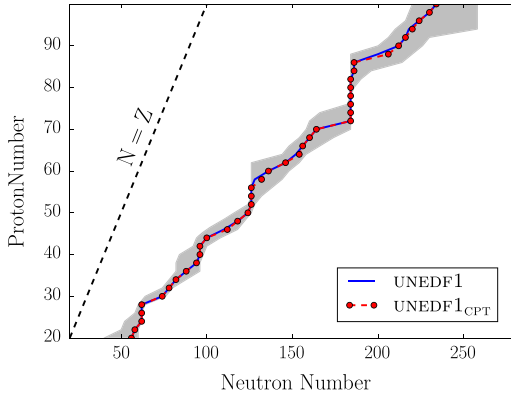


FIG. 3 (color online). Comparison between the two-neutron dripline predictions made with UNEDF1 (solid line) and those made with UNEDF1_{CPT} (dashed line). The 90% probability spread about the UNEDF1 predictions is shown in gray.

producing prediction intervals for the new CPT mass measurements. These estimates are genuine *holdout* predictions since the new mass data were not used in determining the posterior distribution. Figure 2 shows 90% prediction intervals (centered on the mean mass value of UNEDF1) for the new CPT masses. The dark blue band is the 90% interval for the uncertainty due to the EDF model parameters \mathbf{x} alone; the light blue band also includes additional uncertainty due the terms ε_{ij} . The standard deviations s_i were estimated from the difference between the posterior mean estimate and the available mass measurements. Separate estimates were made for mass predictions of spherical and deformed nuclei. These estimated misfit-error standard deviations s_i were assumed to be appropriate for the new CPT mass predictions, producing this additional uncertainty. We observe that the experimentally measured values (black bars in Fig. 2) are generally

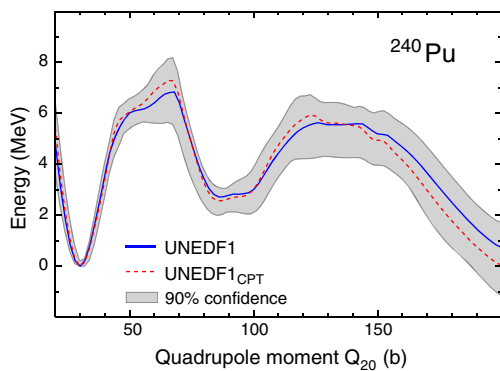


FIG. 4 (color online). Comparison between the fission barrier predictions for ^{240}Pu made with UNEDF1 (solid line) and those made with UNEDF1_{CPT} (dashed line), together with the 90% confidence interval (shaded gray area). The potential energy surface was obtained by following the lowest-energy static fission pathway in a four-dimensional collective space of axial and triaxial quadrupole, axial octupole, and axial hexadecapole mass moments.

within the 90% prediction interval. The estimated uncertainty for the calculated masses is approximately ± 2 MeV, and slightly larger for the four spherical nuclei (the first four nuclei in the figure). This uncertainty is relatively large and in excellent agreement with the rms deviation for masses of even-even nuclei across the entire nuclear landscape, which is 1.9 MeV for UNEDF1.

We now evaluate how the calculated model uncertainties impact predictions for important physical observables. We first look at the position of the two-neutron dripline, which is especially important for our understanding of nucleosynthesis in the r process [40]. For a given element characterized by its proton number Z , the two-neutron dripline is defined as the point where the two-neutron separation energy becomes negative. We have performed an ensemble of calculations of nuclear binding energies for all even-even neutron-rich elements with $20 \leq Z \leq 100$ over the same random sample of EDF parameter inputs, allowing yet another GP-based emulator to be constructed for these binding energies.

Once the emulator is constructed, we propagate the posterior distribution of the model parameters (conditioning on either the UNEDF1 or UNEDF1_{CPT} data sets), producing uncertainty in the estimated dripline. With this Monte Carlo sample, we can estimate the posterior mode and 90% interval for the dripline for each value of Z . We explored the axial quadrupole potential energy surface of each nucleus to allow for deformed solutions. The results are presented in Fig. 3. We observe that the inclusion of 17 new masses of neutron-rich nuclei in the optimization protocol did not impact the position of the dripline, since results with UNEDF1 and UNEDF1_{CPT} are practically indistinguishable. The predicted dripline is consistent with the results of large-scale DFT surveys [6,7]. Apart from the few closed-shell, waiting-point nuclei, the uncertainty on the position of the dripline is on the order of 15 to 20 nucleons. This is comparable to statistical and systematic uncertainties obtained by comparing predictions made with different Skyrme functionals [6].

Another important application area of nuclear DFT is fission theory. In Fig. 4, we show the potential energy curve of ^{240}Pu . This nucleus is representative of the actinide region and is often used as a theoretical benchmark. Again, the results of UNEDF1 and UNEDF1_{CPT} are close. The large theoretical uncertainty in the predicted static fission barrier is worth noting; similar results were obtained in Ref. [41] in the context of fission properties for r -process nuclei. Since a 1 MeV shift in the fission barrier translates into many orders of magnitude difference in the spontaneous fission half-life, such results highlight the urgent need for better constraining the deformation properties of current EDFs.

Conclusions.—We have presented a comprehensive application of Bayesian inference techniques to the calculation and propagation of theoretical statistical uncertainties in nuclear density functional theory. By using the recent,

unique data set of mass measurements from the CPT at Argonne National Laboratory, we showcase how the statistical tools of uncertainty quantification and high-performance computing can be used to assess the information content of new data with respect to current models. Such analyses will become increasingly relevant for enhancing the feedback in the “observation-theory-prediction-experiment”—cycle of the scientific method at the eve of next-generation radioactive ion beam facilities and exascale computing.

In the particular case studied in this work, we found that the impact of the new neutron-rich nuclei mass data on our DFT model is minor. The coupling constants of the earlier functional UNEDF1 and of the new functional UNEDF1_{CPT}, informed by the new data, are fairly close; hence, their predictions for the two-neutron dripline and fission barrier in ²⁴⁰Pu are practically identical. Although the major theoretical statistical uncertainty in developments of the nuclear EDF comes from the poorly constrained isovector terms and the new data on neutron-rich nuclei are generally expected to reduce this uncertainty, the lack of a significant constraint from the new masses suggests that both the amount of new neutron-rich isotope data and the range of neutron excess probed are not sufficiently large to impact our model appreciably. Moreover, because of their poor precision with respect to the existing data (see Table I), even the current, best-calibrated EDFs are not sensitive and flexible enough to fully take advantage of the new experimental information.

By propagating theoretical errors, we found large model uncertainties in the predictions of the two-neutron dripline and the fission barrier in ²⁴⁰Pu. In this respect, we concur with the conclusions of Ref. [30] that existing mass models are insufficient for accurate *r*-process simulations. Clearly, accurate measurements for nuclei with even larger neutron excess, closer to the *r*-process path, are still needed in order to better inform theory.

We note that the uncertainties discussed in this work are estimated statistically, reflecting parameter uncertainty and model misfit. The misfit error is most likely due to our lack of knowledge of the form of the nuclear EDF itself, and additional measurements will never reduce this source of uncertainty. Adding physics that is missing in the current implementations of nuclear DFT is a major challenge for the field. A distinct and complementary challenge is to develop tools that deliver uncertainty quantification for theoretical studies as well as for the assessment of new experimental data. The present work represents a step in this direction.

We are grateful to Guy Savard and Robert Janssens for helpful discussions and access to CPT masses. This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics under Awards No. DE-AC52-07NA27344 (Lawrence Livermore National Laboratory),

No. DE-AC02-06CH11357 (Argonne National Laboratory), and No. DE-SC0008511 (NUCLEI SciDAC Collaboration), and by the NNSA’s Stewardship Science Academic Alliances Program under Grant No. DE-NA0001820. Computational resources were provided through an INCITE award “Computational Nuclear Structure” by the National Center for Computational Sciences and National Institute for Computational Sciences at Oak Ridge National Laboratory, through a grant by the Livermore Computing Resource Center at Lawrence Livermore National Laboratory, and through a grant by the Laboratory Computing Resource Center at Argonne National Laboratory.

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