A Bayesian Approach for Parameter Estimation and Prediction using a DFT Model for Binding Energies

Dave Higdon, Jordan McDonnell, Nicolas Schunck, Jason Sarich, Stefan Wild, Witek Nazarewicz

LANL LLNL LLNL ANL ANL UTK

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NUCLEI Nuclear Computational Low-Energy Initiative

Los Alamos NATIONAL LABORATORY EST. 1943

Abstract

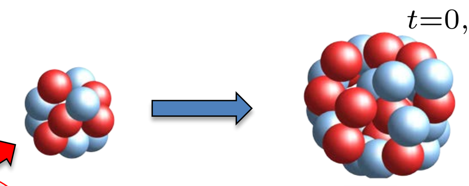
Bayesian methods have been very successful in quantifying uncertainty in physics-based problems in parameter estimation and prediction. In these cases, physical measurements y are modeled as the best fit of a physics-based model η(θ), where θ denotes the uncertain, best input setting. Hence the statistical model is of the form

$$y = \eta(\theta) + \varepsilon,$$

where ε accounts for measurement, and possibly other error sources. When non- linearity is present in η(·), the resulting posterior distribution for the unknown parameters in the Bayesian formulation is typically complex and non-standard, requiring computationally demanding computational approaches such as Markov chain Monte Carlo (MCMC) to produce multivariate draws from the posterior. While quite generally applicable, MCMC requires thousands, or even millions of evaluations of the physics model η(·). This is problematic if the model takes hours or days to evaluate. To overcome this computational bottleneck, we present an approach adapted from Bayesian model calibration. This approach combines output from an ensemble of computational model runs with physical measurements, within a statistical formulation, to carry out inference. A key component of this approach is a statistical response surface, or emulator, estimated from the ensemble of model runs. We demonstrate this approach with a case study in estimating parameters for a density functional theory (DFT) model, using experimental measurements from a collection of atomic nuclei. We also demonstrate how this approach produces uncertainties in predictions for recent mass measurements obtained at CARIBU Facility at Argonne National Laboratory (ANL).

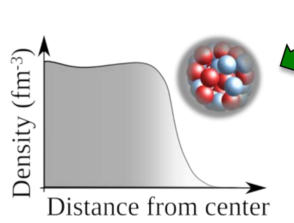
Nuclear Density Functional Theory

$$E = \int \left[\mathcal{E}_{\rm kin}(\boldsymbol{r}) + \mathcal{E}_{\rm Cou}(\boldsymbol{r}) + \mathcal{E}_{\rm pair}(\boldsymbol{r}) + \sum_{t=0,1} \chi_t(\boldsymbol{r}) \right] d^3\boldsymbol{r}$$

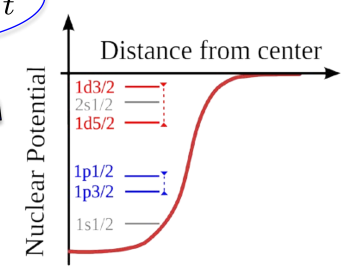


$$\chi_t(\boldsymbol{r}) = C_t^{\rho\rho}\rho_t^2 + C_t^{\rho\tau}\rho_t\tau_t + C_t^{JJ}\sum_{\mu\nu} J_{\mu\nu,t}J_{\mu\nu,t} + C_t^{\rho\Delta\rho}\rho_t\Delta\rho_t + C_t^{\rho\nabla J}\rho_t\nabla\cdot J_t$$

- Built on effective nuclear forces between protons and neutrons
- Uses densities of nucleons as main building blocks

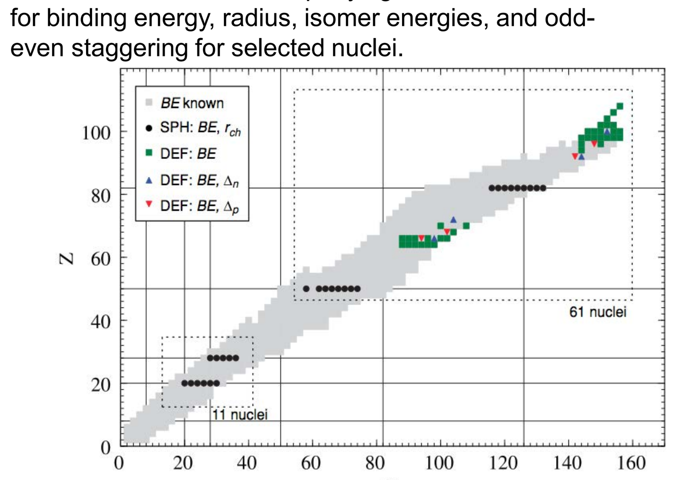


DFT solver determines ρ, τ, J for specified parameters

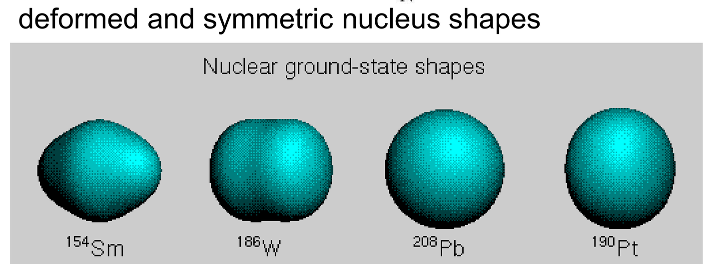


Measurements and Quantities of Interest

Measurements and accompanying simulations are made for binding energy, radius, isomer energies, and odd-even staggering for selected nuclei.

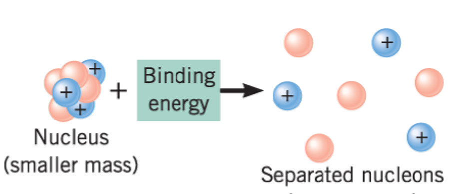


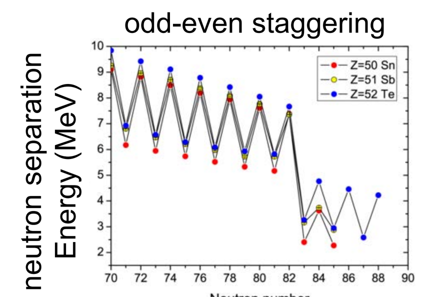
deformed and symmetric nucleus shapes



Charge radius can be measured for symmetric nuclei
Shapes obtained from DFT code

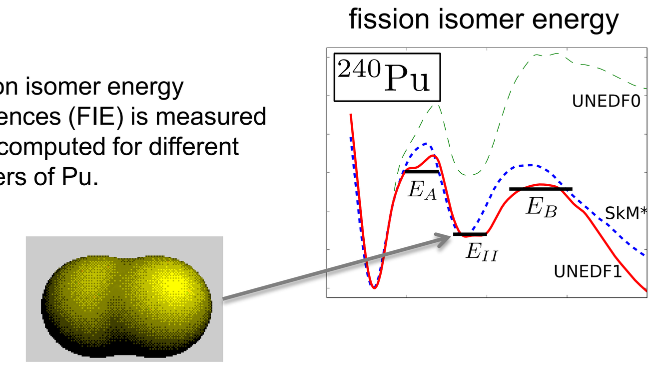
Binding energy is the additional energy released when a nucleus is completely separated.



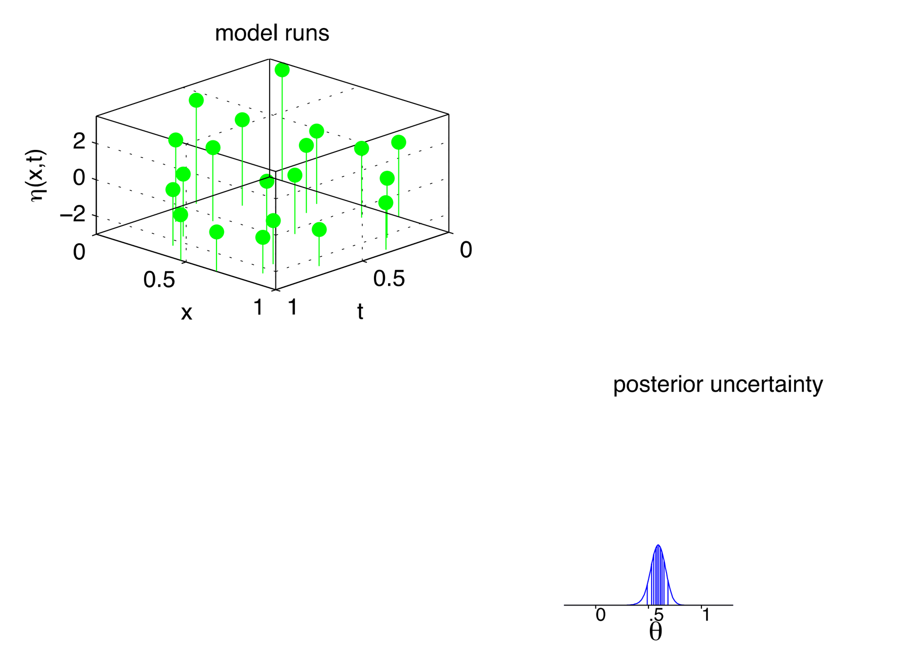


Odd-even staggering measures the difference in binding energy between odd and even nuclei. This is computed for ±1 neutron (OESn) or proton (OESp)

Fission isomer energy differences (FIE) is measured (and computed for different isomers of Pu.



Bayesian Model Calibration Framework is Used to Combine Simulation Output and Experiments to Estimate Model Parameters and Make Predictions



Simulations and experimental observations are combined according the the Bayesian model calibration of Kennedy and O'Hagan (2001).

- x model or system inputs
- θ model calibration parameters
- $\zeta(x)$ true physical system response given inputs x
- $\eta(x,\theta)$ simulator response at x and θ.
- $y(x)$ experimental observation of the physical system
- $\delta(x)$ discrepancy between $\zeta(x)$ and $\eta(x,\theta)$ may be decomposed into numerical error and bias
- $e(x)$ observation error of the experimental data

$$y(x) = \zeta(x) + e(x)$$
$$y(x) = \eta(x,\theta) + \delta(x) + e(x)$$

References:

M. Kennedy and A. O'Hagan. Bayesian calibration of computer models (with discussion). Journal of the Royal Statistical Society (Series B), 68:425–464, 2001.

D. Higdon, J. Gattiker, B. Williams, and M. Rightley. Computer Model Calibration Using High-Dimensional Output. Journal of the American Statistical Association, 103(482):570–583, 2008.

D. Higdon, J. McDonnell, N. Schunck, S. Sarich, S.M. Wild. A Bayesian approach for parameter estimaton and prediction using a computationally intensive model. 2014.

Bayesian Model Formulation

$$y_{ij} = \eta(\theta, i, j) + \delta_{ij} + \epsilon_{ij}$$

misfit experimental error

Measurement model: $e_{ij} = \delta_{ij} + \epsilon_{ij} \sim N(0, \lambda_i^{-1})$

$$L(y|\theta,\lambda) \propto \prod_{i,j} \lambda_i^{\frac{1}{2}} \exp\left\{-\frac{1}{2}\lambda_i(y_{ij} - \eta(\theta,i,j))^2\right\}$$

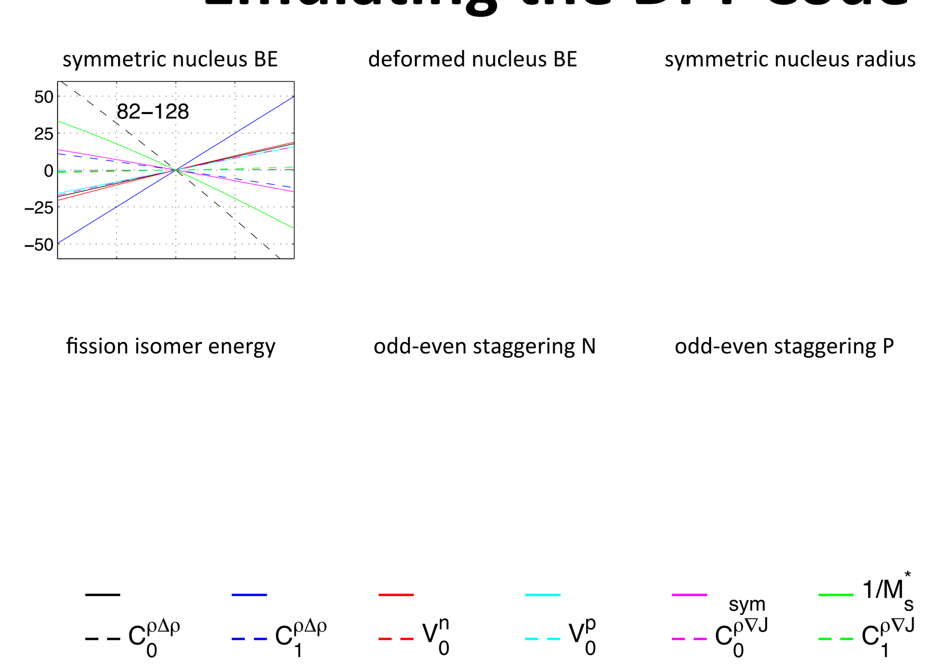
Prior model

$$\pi(\theta) \propto I[\theta \in \mathcal{S}]$$
$$\pi(\lambda_i) \propto \lambda_i^{a-1}e^{-b\lambda_i}$$
$$\eta(\cdot) \sim GP\left(\mu(\cdot), C_\beta(\cdot,\cdot)\right)$$
$$\pi(\beta)$$

Posterior

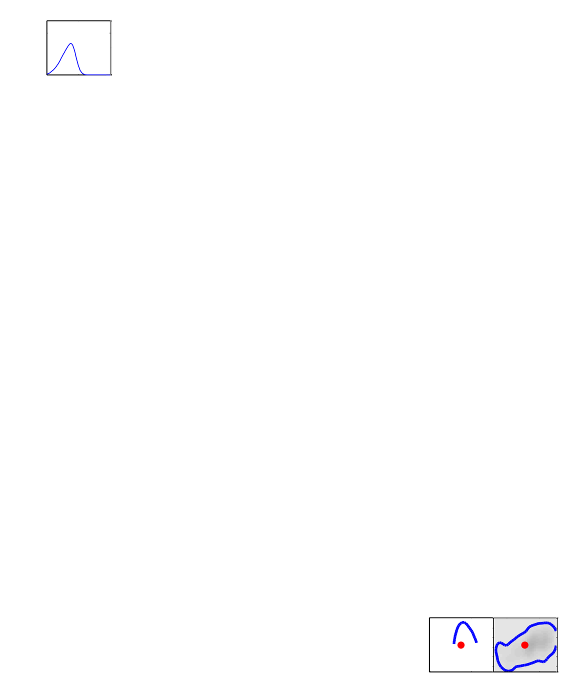
$$\pi(\theta, \lambda, \eta(\cdot), \beta) \propto L(y|\theta,\lambda) \times \pi(\theta) \times \pi(\lambda) \times \pi(\eta(\cdot)) \times \pi(\beta)$$

Emulating the DFT Code & Sensitivities



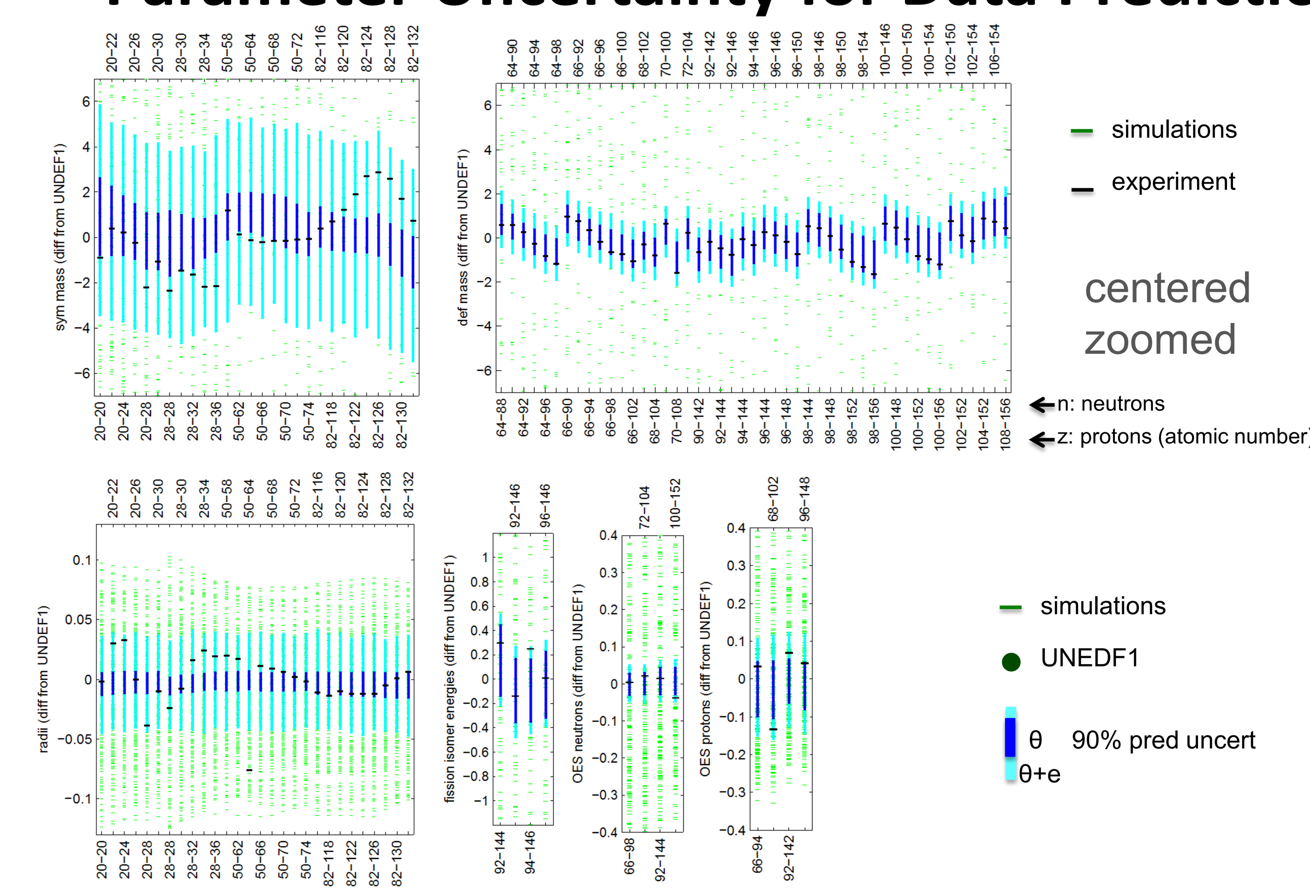
Estimating the response surface emulator η(•) allows exploration of the behavior of the DFT code as input parameters are varied. This figure shows the sensitivity of the masses computed via DFT as each of the 12 parameters are varied from low to high. The plots show how computed masses change for three different nuclei (Z-N) – one spherical, one deformed, and one from the new ANL measurements – as the parameters are varied, one at at time.

Posterior Uncertainty for DFT Parameters θ

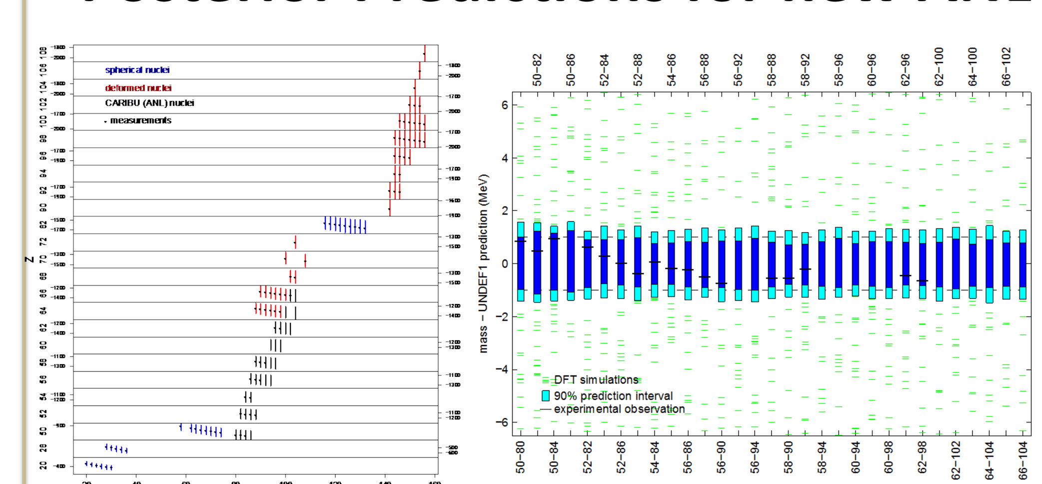


Posterior uncertainty for the 12-d DFT parameter vector after conditioning on the various data sources: masses (spherical nuclei), masses (deformed nuclei), charge radius (spherical radius), fission isomer energy, odd-even staggering (neutrons), odd-even staggering (protons). 1-d densities are shown along the diagonal of the figure; 2-d marginal distributions are represented (redundantly) along the off-diagonals.

Parameter Uncertainty for Data Predictions

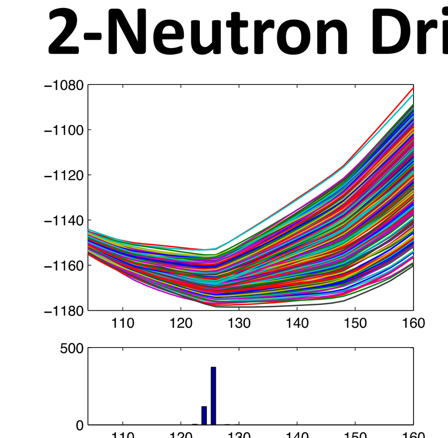


Posterior Predictions for new ANL N-rich Measurements

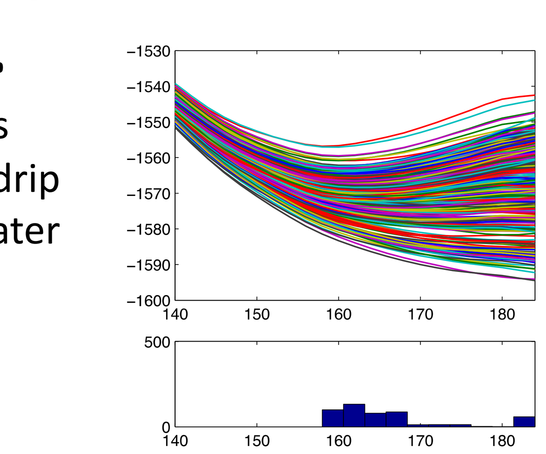


Predictions for recently produced mass measurements taken at ANL's CARIBU facility. Unlike the above predictions, these predictions are not for measurements that were used to estimate the model parameters. These holdout predictions agree very well with the actual measurements.

Parameter Uncertainty Propagated thru to Estimate 2-Neutron Drip Probabilities.



Neutron drip probabilities for Tin and Yb. Here the drip line uncertainty is far greater for Yb.



Drip probabilities are estimated from Monte Carlo simulations of binding energy curves for each Z