Uncertainty assessment for calculated atomic, molecular and nuclear data and implications for GUM

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Editorial Statement, *Phys Rev A* (2011)

Papers presenting the results of theoretical calculations are expected to include uncertainty estimates for the calculations whenever practicable, and especially under the following circumstances:

- If the authors claim high accuracy, or improvements on the accuracy of previous work.
- If the primary motivation for the paper is to make comparisons with present or future high precision experimental measurements.
- If the primary motivation is to provide interpolations or extrapolations of known experimental measurements.

In practice: Policy works well for calculated structure data, not so well for calculated scattering data. (For structure data spectroscopy provides experimental benchmarks.)

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Calculated data, classes of computations

Point of terminology: It is Calculated or Measured data (or both) according to where is the main source of uncertainty.

Three main classes of computations:

- Applied numerical analysis: precisely specified problems typically belonging to linear algebra, optimization and approximation, differential equations, integral equations and to some extent stochastic systems.
- Simulation of complex systems: basic equations may not be well established, may involve poorly known parameters and functional dependencies, include stochastic elements and may give rise to chaotic behaviour.
- Calculations for simple systems that are computationally hard: prime examples are electronic structure and other many-body quantum mechanics; also problems in combinatorial optimization.

Uncertainty Analysis varies over these three classes.

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Variable status of uncertainty assessment

Uncertainty assessment for the 3 classes:

- Applied numerical analysis: There is the concept of an exact value and of convergence of the numerical method and there is a theory of discretization error and rounding error; this is the core of classical numerical analysis. It has no need for guidance from VIM and GUM.
- Simulation of complex systems: The uncertainty analysis is the domain of Uncertainty Quantification (UQ); note SIAM and GAMM activity groups, meetings, NAS report, thrust area for support. Concern with uncertainty propagation for stochastic systems; “polynomial chaos.” The field of UQ can provide examples for GUM.
- Simple systems that are computationally hard: Science of uncertainty assessment or UQ needs to be developed for specific applications.

IAEA Atomic and Molecular (A+M) Data Unit is concerned with uncertainty assessment for calculated A+M data. This is all based on electronic structure theory; simple systems that are computationally hard.

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Nature of the Quantity of Interest (QoI)

Often high-dimensional. A function rather than a number; e.g. a cross section as a function of energy; a differential cross section; an equation of state. In that case, uncertainties in the components of the QoI are strongly correlated.

Sometimes also measured. In atomic spectra: line positions and amplitudes.

Often very hard or impossible to measure. State-specific cross sections; equation of state under extreme conditions.

Sometimes doesn't make sense as a measured quantity. The Schrödinger wavefunction. Climate sensitivity.

Nature of the calculation

First principles calculations: In general it means, no tunable parameters. In the context of A+M physics it means calculations based on the many-body Schrödinger equation. Mainly relevant for atoms and small molecules.

Less than first principles: Tunable parameters, but experiment is in the background (e.g., parameters have been tuned in some transferable way). Density functional theory (DFT) for large molecules and condensed phase; general force fields.

Based on models: Calibrated to experimental data. Includes all of nuclear physics: structure, decay, scattering. (Lattice QCD would be first principles.)

Contribution from IAEA A+M Data Unit

Our task: To provide internationally recommended and validated data for A+M+PMI/PSI processes relevant to fusion.

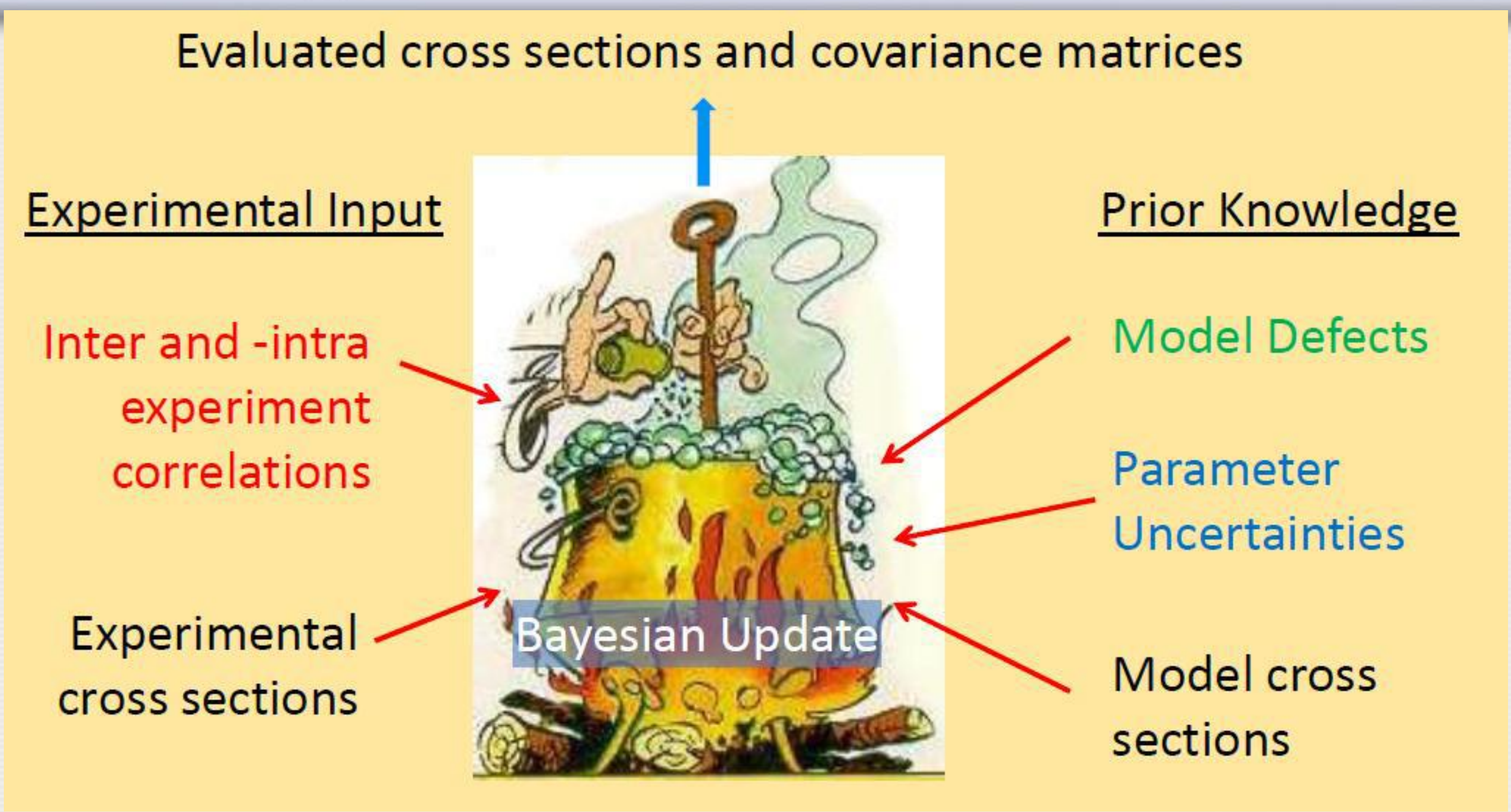
Challenge: Develop practical methods to estimate uncertainties of calculated data.

Meeting highlight: IAEA-ITAMP Technical Meeting on Uncertainty Assessment for Theoretical Atomic and Molecular Scattering Data, Cambridge, MA, 7-9 July 2014.

This presentation: One approach from the nuclear data community; Unified Monte Carlo.

Call for a new discipline: Uncertainty Quantification for simple physical systems that are computationally hard.

Unified Monte Carlo Approach for Nuclear Data



From D. Neudecker, S. Gundacker, H. Leeb et al., ND2010, Jeju Island, Korea;
Via R. Capote, presentation at IAEA, 2013-05-06

Unified Monte Carlo Approach for Nuclear Data

Following R. Capote, presentation at IAEA, 2013-05-06

- $p(\sigma) = C \text{ x } \mathscr{L}(y_E,V_E \mid \sigma) \text{ x } p_0(\sigma \mid \sigma_C,V_C)$
- $p_0(\sigma \mid \sigma_C,V_C) \sim \exp\{-(½)[(\sigma-\sigma_C)^T \bullet (V_C)^{-1} \bullet (\sigma-\sigma_C)]\}$
- $\mathscr{L}(y_E,V_E \mid \sigma) \sim \exp\{-(½)[(y-y_E)^T \bullet(V_E)^{-1} \bullet (y-y_E)]\}, y=f(\sigma)$
- y_E, V_E: measured quantities with *n* elements
- σ_C, V_C: calculated using nuclear models with *m* elements

Use Metropolis (Markov chain) sampling for **σ**.

[] D. L. Smith, “A Unified Monte Carlo Approach to Fast Neutron Cross Section Data Evaluation,” Proceedings of the 8th International Topical Meeting on Nuclear Applications and Utilization of Accelerators, Pocatello, Jul 29 – Aug 2 2007, p. 736.

[] R. Capote and D. L. Smith, “Unified Monte Carlo and Mixed Probability Functions,” Journal of the Korean Physical Society 59 (2), August 2011, pp. 1284-1287 (Proceedings ND2010).

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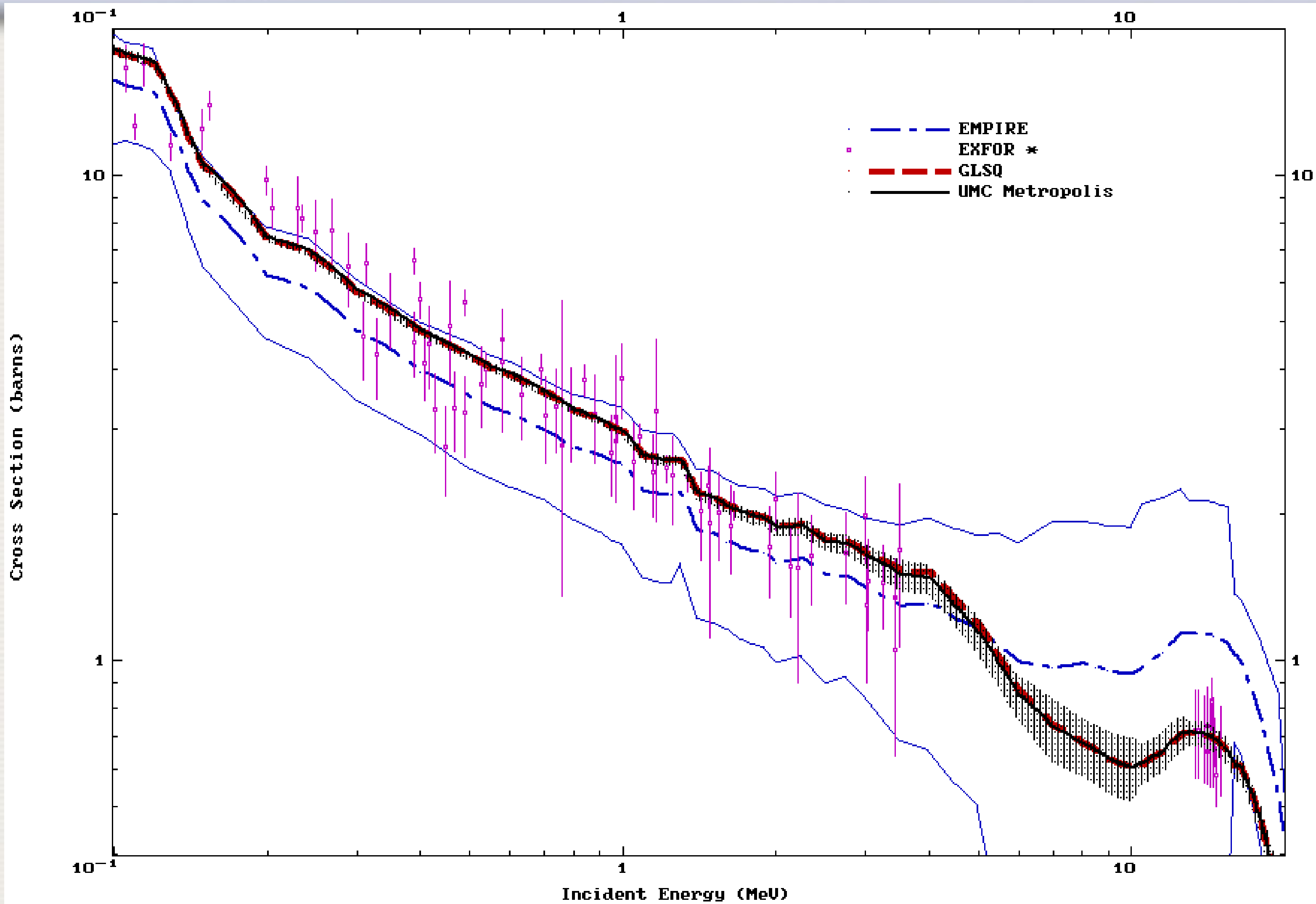
Unified Monte Carlo Approach for Nuclear Data



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Summary remarks: numerical analysis vs UQ

Numerical Analysis deals with precisely specified problems.

- There is a concept of an exact answer.
- One studies discretization error, truncation error.

Electronic structure has NA aspects.

- (one electron) basis set extrapolation.
- R-matrix convergence wrt radius.

Complexity of electronic structure goes beyond NA.

- Cannot extrapolate to Full CI limit.
- Must rely on models, e.g. DFT.
- Most difficult: electronic excitation and condensed matter.

Challenge for future work to expand domain of UQ

Summary remarks: calculated data in GUM

In the present GUM: Uncertainties in calculated data are basically out of scope. If calculations can be described as experiments then elements of GUM could apply, but examples are not developed.

In a revised GUM: Questionable.

I don't expect that a practitioner that uses tools from numerical analysis will look to GUM-rev for the uncertainty assessment.

Probably GUM-rev can benefit from a close look at UQ and use it as a source of examples. Probably GUM-rev should refer to tools of UQ; beyond that, I don't know.

The uncertainty assessment for computationally hard simple systems needs science development.