Pacific Northwest
NATIONAL LABORATORY

The Computational Chemical Sciences (CCS) Program at BES and PNNL:

Present status and ideas for the near future

July 27, 2023

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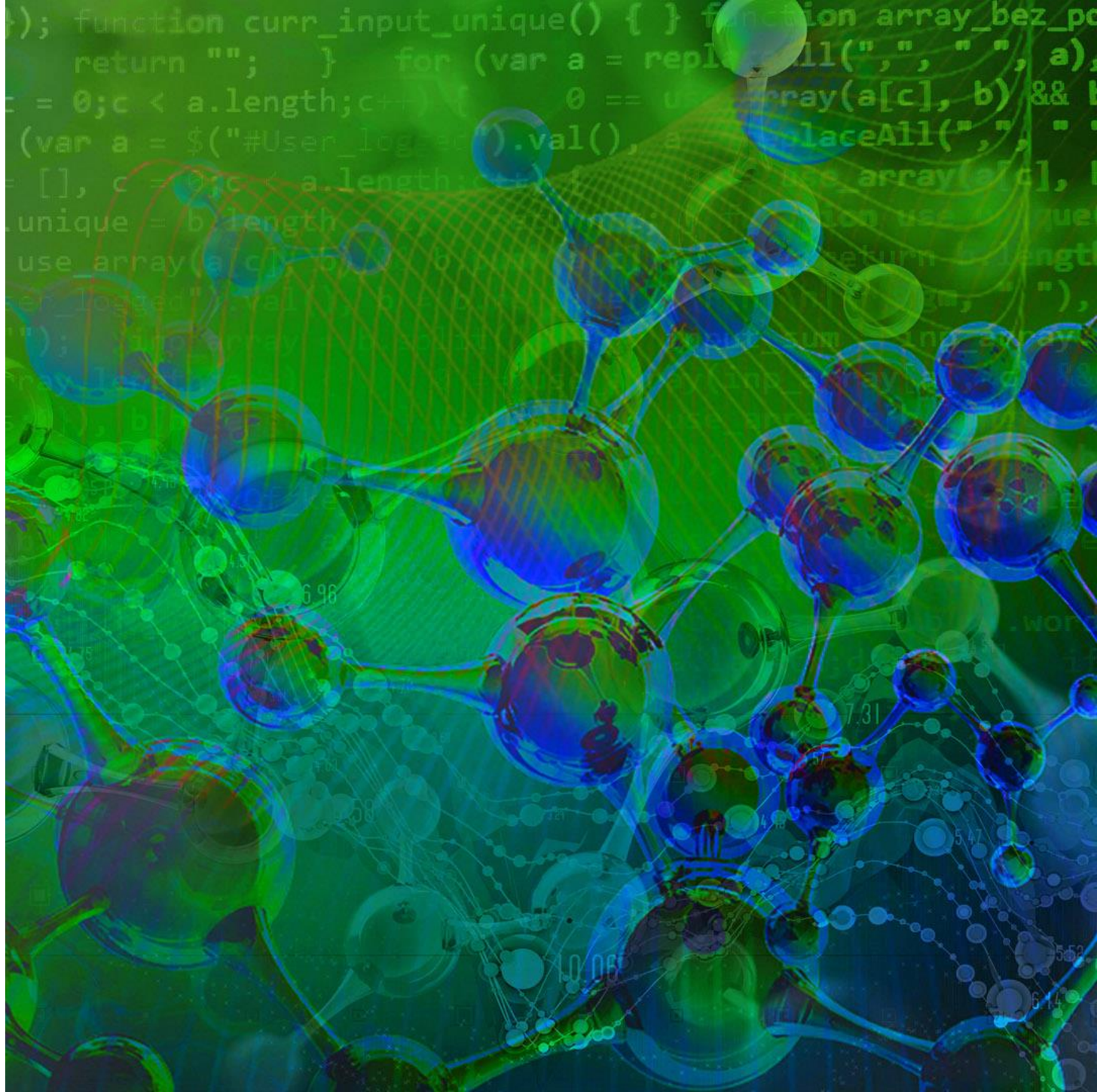
Pacific Northwest National Laboratory
University of Washington



U.S. DEPARTMENT OF ENERGY BATTELLE

PNNL is operated by Battelle for the U.S. Department of Energy

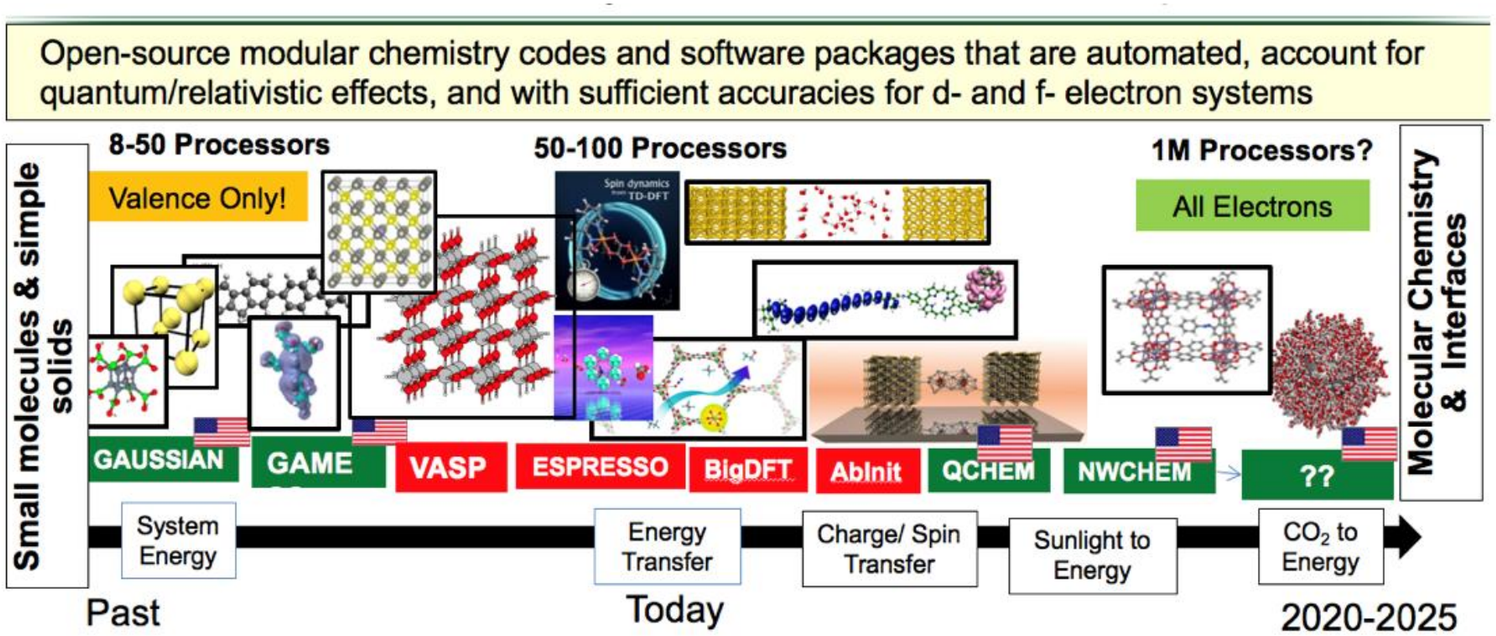
Path to the Computational Chemical Sciences Program

BES recognized the importance of a new electronic structure software that is:

- US-developed
- Open source
- Scalable

The software should include:

- Efficient scaling on ~1M processors
- Relativistic effects
- Proper inclusion of spin (proper treatment of d- and f- electrons)
- Improvements on DFT



Quantum effects at surfaces, interfaces, solutions, and vacuum govern chemical energy conversion, & electron transport in molecular-scale devices. **Spin-dependent effects must be included for**

- Artificial photosynthesis
- Gas separation and conversion
- Molecular catalysts
- Next generation computing chips

US must continue to lead development of high-accuracy chemical software that …

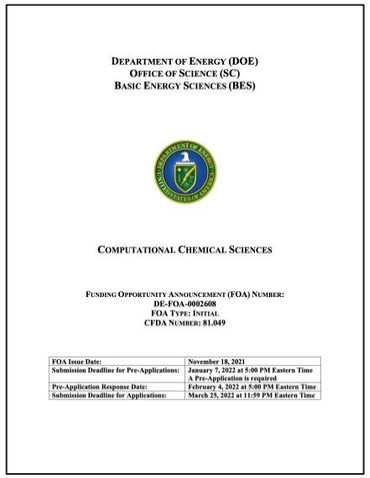
- Is compatible with low memory peta-to-exascale computing architectures
- Achieve efficient scaling of quantum methods with the number of atoms
- Accounts for relativistic effects in f-electrons
- Improves on DFT methods (self-interaction corrections, accuracy, new basis sets)

Harriet Kung

BESAC Presentation, February 2016
https://science.osti.gov/bes/besac/Meetings/Meeting-Presentations/201602

Path to the Computational Chemical Sciences Program

- Performance from advances in processor speed has been nearly exhausted
- Performance and scaling must come from parallelization (software)

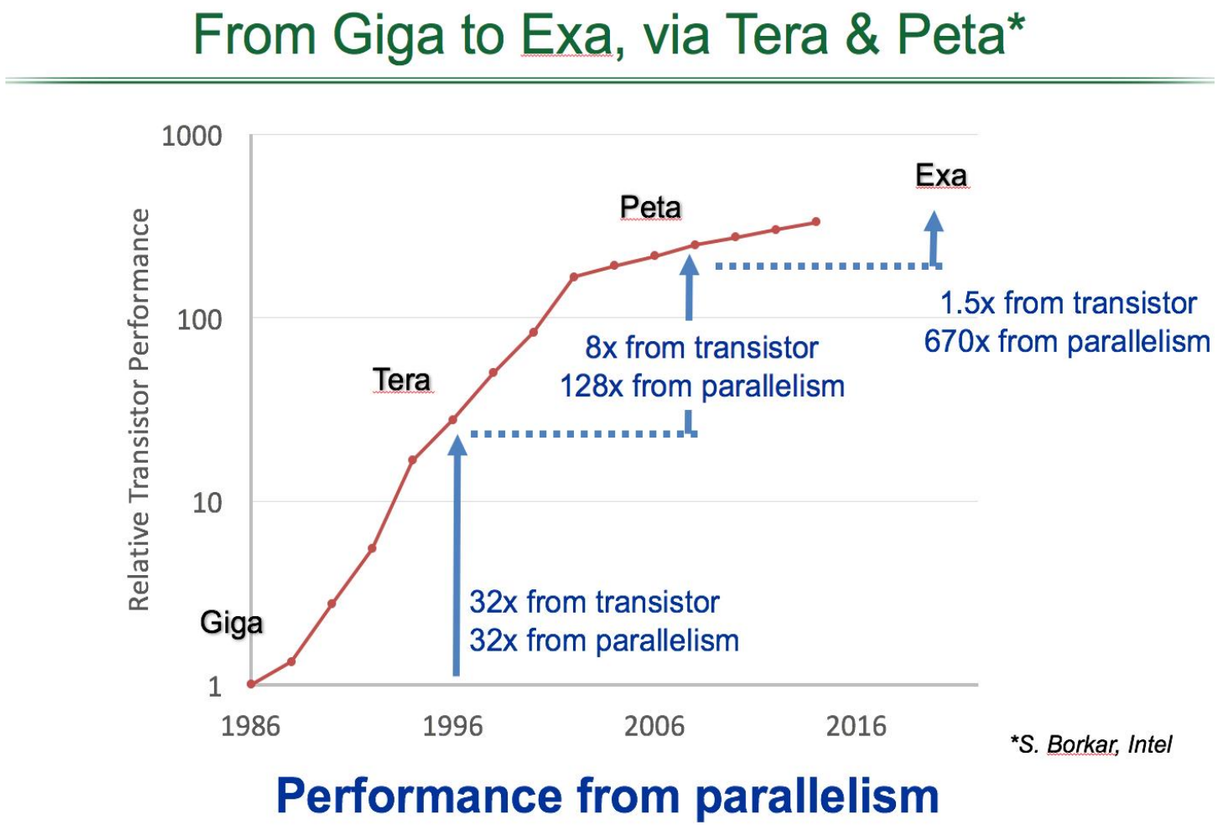


DEPARTMENT OF ENERGY (DOE)
OFFICE OF SCIENCE (SC)
BASIC ENERGY SCIENCES (BES)

COMPUTATIONAL CHEMICAL SCIENCES

FUNDING OPPORTUNITY ANNOUNCEMENT (FOA) NUMBER:
DE-FOA-0002608
FOA TYPE: INITIAL
CFDA NUMBER: 81.049

FOA Issue Date:	November 18, 2021
Submission Deadline for Pre-Applications:	January 7, 2022 at 5:00 PM Eastern Time A Pre-Application is required
Pre-Application Response Date:	February 4, 2022 at 5:00 PM Eastern Time
Submission Deadline for Applications:	March 25, 2022 at 11:59 PM Eastern Time



Steve Binkley — **BESAC Presentation, February 2016**
https://science.osti.gov/bes/besac/Meetings/Meeting-Presentations/201602

Current Status of the Computational Chemical Sciences Program

Currently 16 Funded Projects

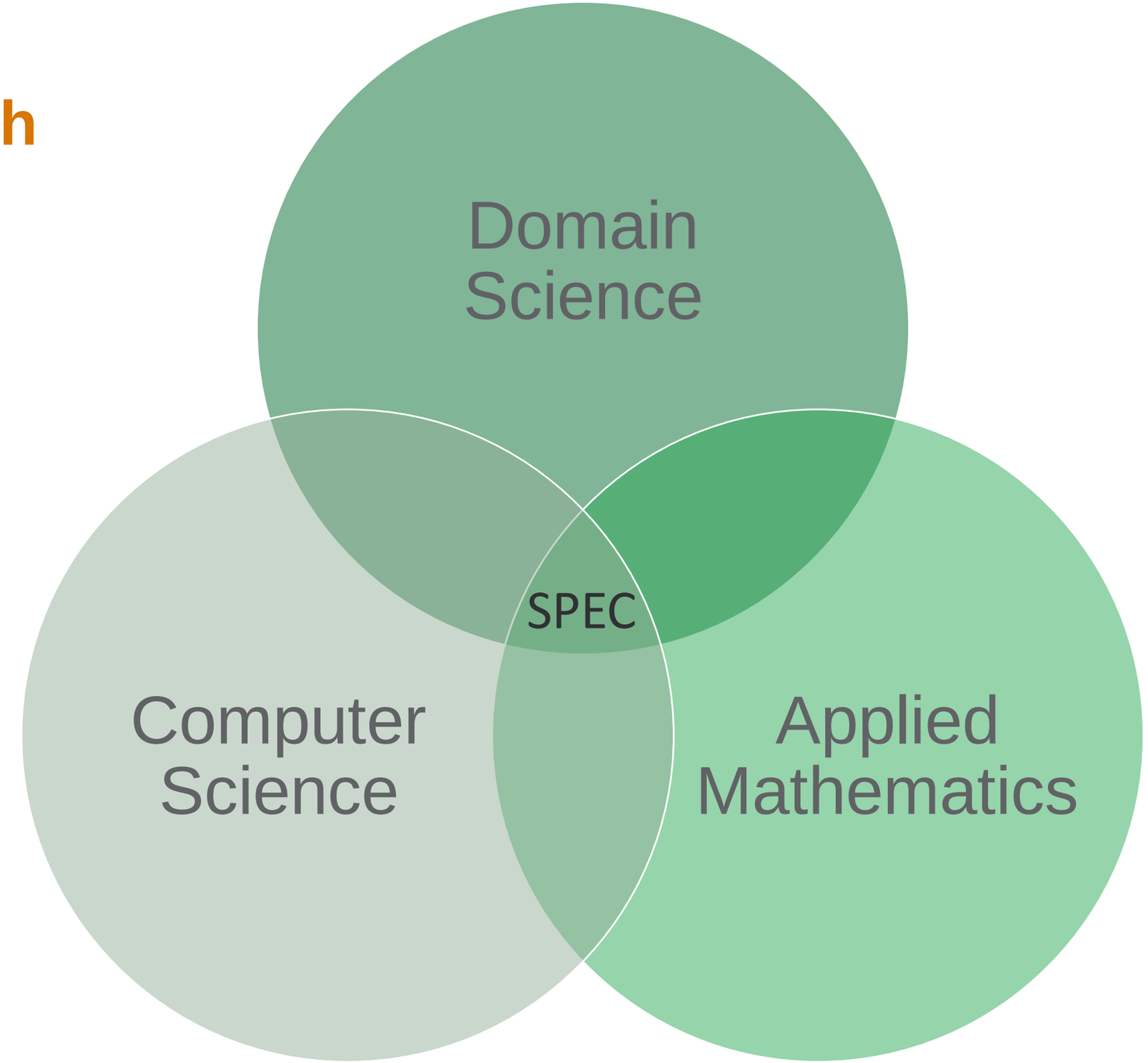
- University of Chicago (2)
- Princeton University
- Georgia Tech
- University of California Santa Cruz
- Yale University
- Central Michigan University
- University of Southern California
- Rensselaer Polytechnic Institute
- Virginia Tech
- University of Michigan
- Pacific Northwest National Laboratory (2)
- Ames Laboratory
- National Renewable Energy Laboratory
- Sandia National Laboratories

Software libraries for previous and current projects are deposited at *https://ccs-psi.org* (University of Minnesota)

Our Approach

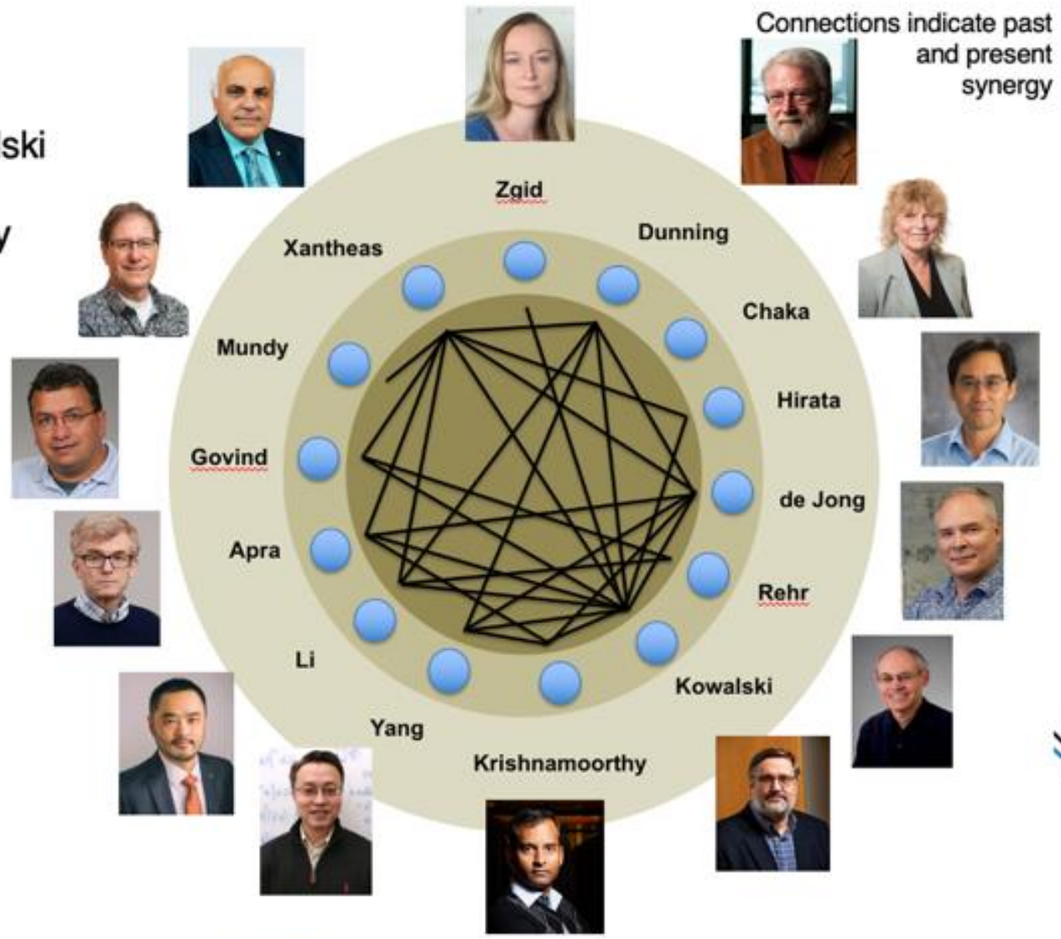
- Implement state-of-the-art, many-body electronic structure methodologies
- Deliver scalable, open-source electronic structure software libraries
- Address challenges in excited-states of complex chemical systems
- Interpret signals obtained at DOE's light source facilities (APS, ALS, LCLS, NSLS)

https://spec.labworks.org



Scalable Predictive methods for Excitations and Correlated phenomena (SPEC)

- **PNNL**
  - Sotiris Xantheas, Karol Kowalski
  - Edoardo Apra, Niri Govind
  - Anne Chaka, Chris Mundy
- **Univ. of Washington**
  - Xiaosong Li
  - Thom Dunning, Jr.
  - John Rehr
- **University of Michigan**
  - Dominika Zgid
- **LBNL**
  - Wibe de Jong
  - Chao Yang
- **University of Illinois**
  - So Hirata

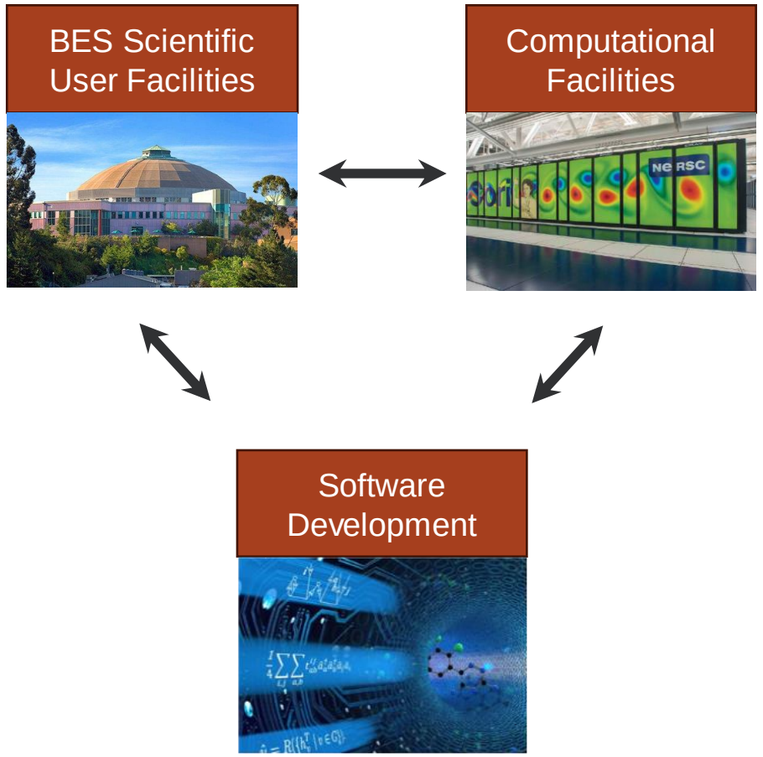


- **J. Heyrovsky Institute**
  - Jiří Brabec (Prague)
  - Libor Veis (Prague)

- **Wigner Institute**
  - Ors Legeza (Budapest)

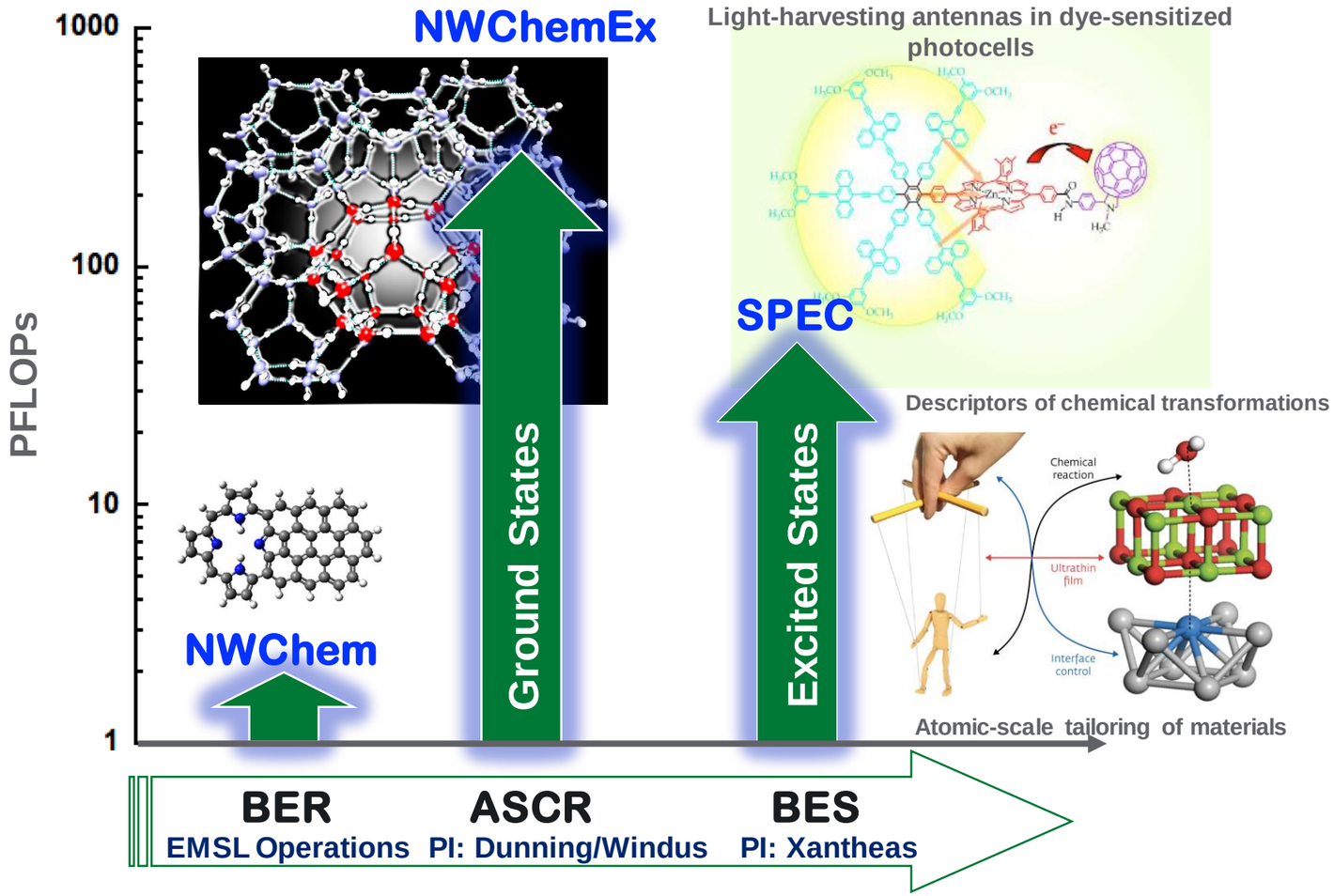
https://spec.labworks.org

Role of Software Development in the DOE Complex

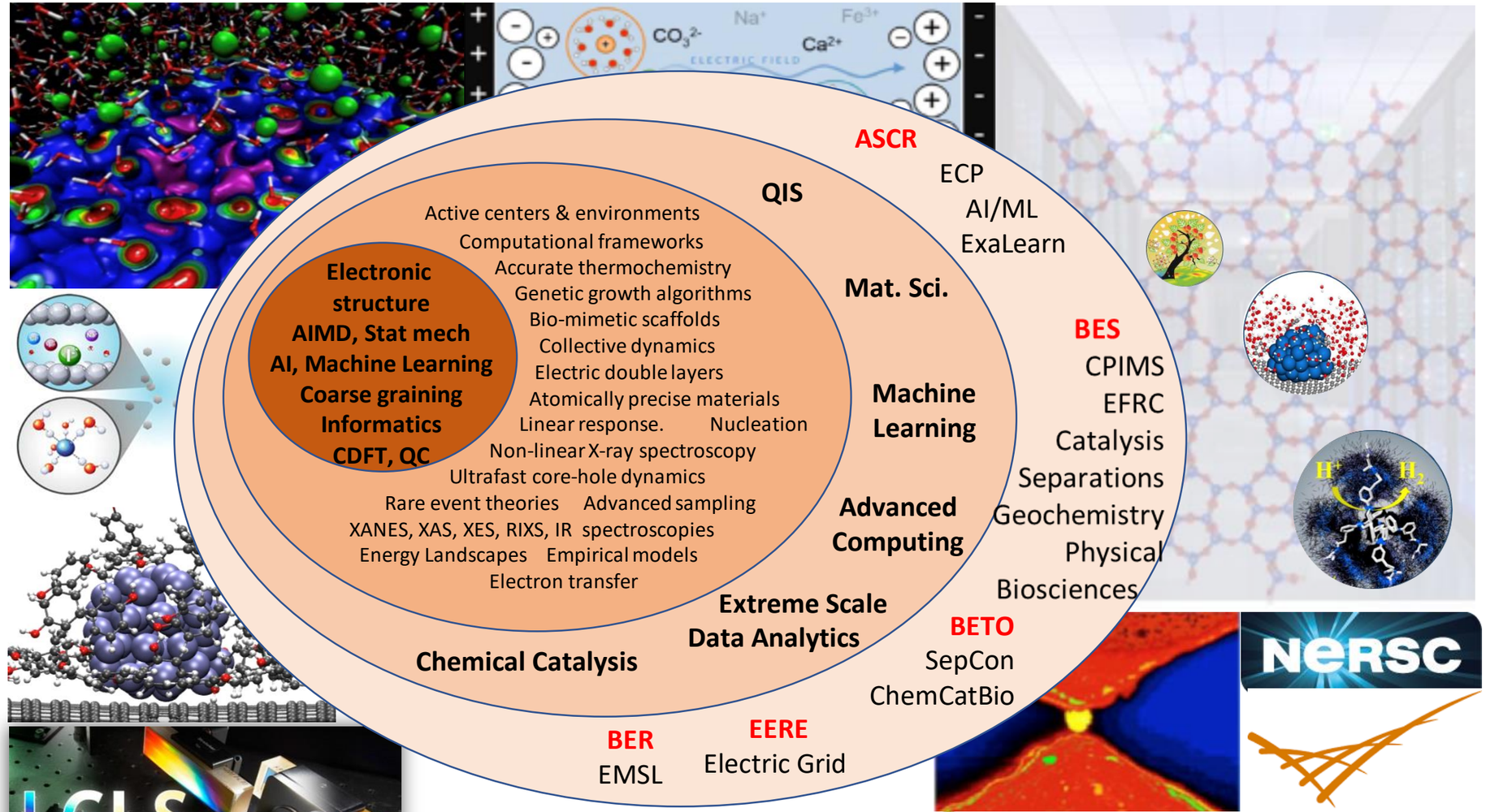


New scientific discoveries enabled by software development

Scalable software development at PNNL



Computational and Theoretical Chemistry Institute
CTCI@PNNL

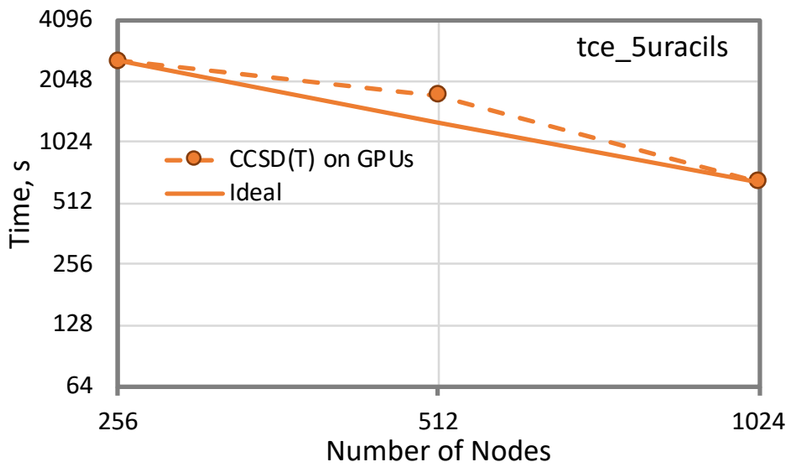


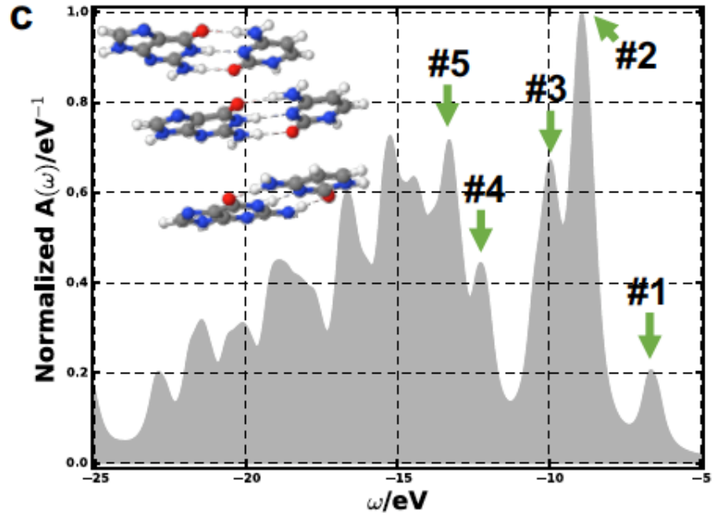
Ecosystem:

- Methods
- Applications
- PNNL Investments
- Sponsors

Scaling SPEC Libraries on Summit using TAMM

SPEC: CCSD(T) on GPUs





Peng, Panyala, Kowalski, Krishnamoorthy, *Comp. Phys. Comm.* **265**, 108000 (2021)

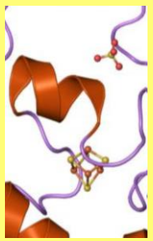
Peng, Kowalski, Panyala, Krishnamoorthy *J. Chem. Phys.* **152**, 011101 (2020)

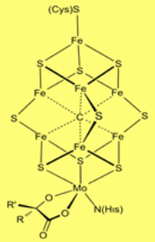
Timings / Scaling (CCSD)

3 base pairs, 1,173 basis functions on 256 nodes (36 cores per node) – 76 min per iteration

Robust, scalable, selected CI implementation for MCSCF

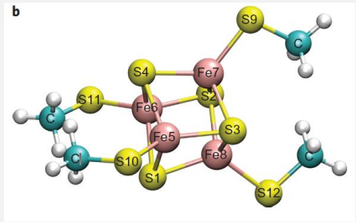
"FeS"





"FeMoco"

Sharma, Sivalingam, Neese and Chan, *Nat. Chem.* **6**, 927 (2014)



Towards modeling active centers in enzymes, *Simone Raugei* (PNNL)

Brabec, Brandejs, Kowalski, Xantheas, Legeza, Veis, *J Comput Chem.* **42**, 534 (2021)

JCTC **18** (2), pp. 687-702 (2022); DOI:10.1021/acs.jctc.1c00830

JCTC **17** (10), pp. 6080-6091 (2021); DOI:10.1021/acs.jctc.1c00485

Looking to the Future

- **Challenges**
  - How do we move from a model to real systems that resemble the ones measured in DOE's light sources?
  - How do we realize the full potential of upgrades at DOE computational facilities (NERSC) and other DOE offices (ASCR)?
  - How do we involve the scientific community in a concerted effort to realize the full potential of developed software?
  - How do we train early career scientists to fully realize the advantages of heterogeneous hardware architectures?
- **Underlying issues**
  - Speed, quantum co-design
  - Scaling
  - Sustainability
- **Path forward**
  - Request for information from ASCR
  - Workshop on sustainable software (November 2022)

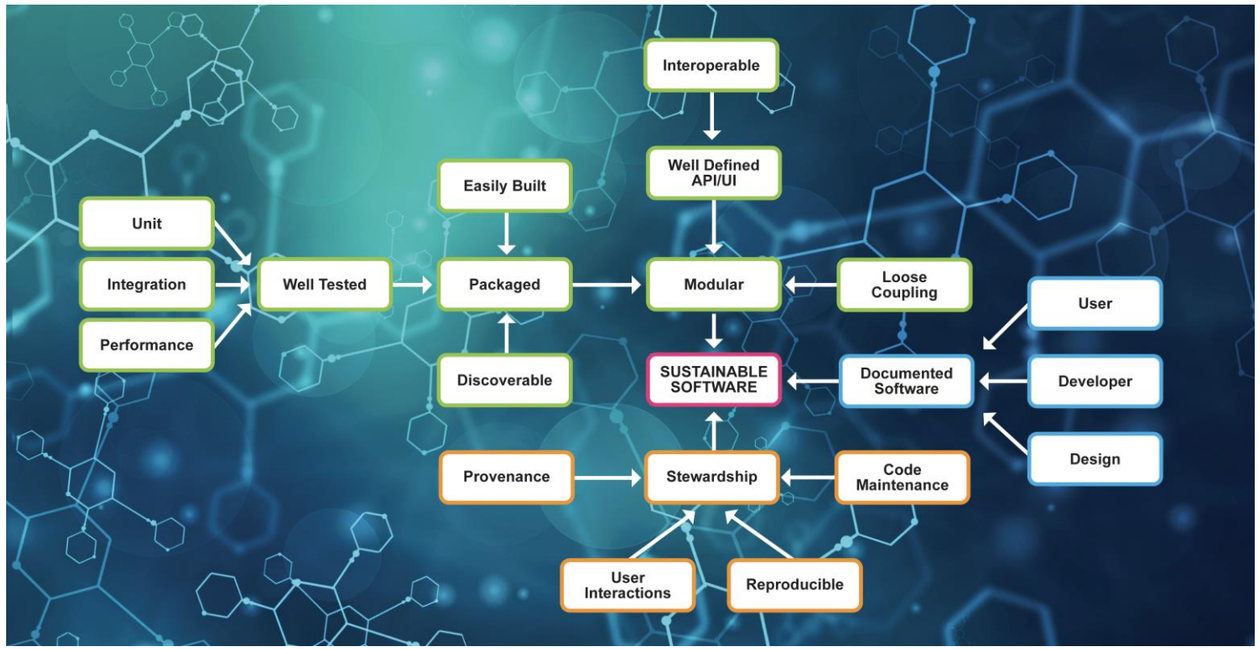
U.S. DEPARTMENT OF ENERGY | Office of Science

Department of Energy Releases Request for Information on Software Stewardship

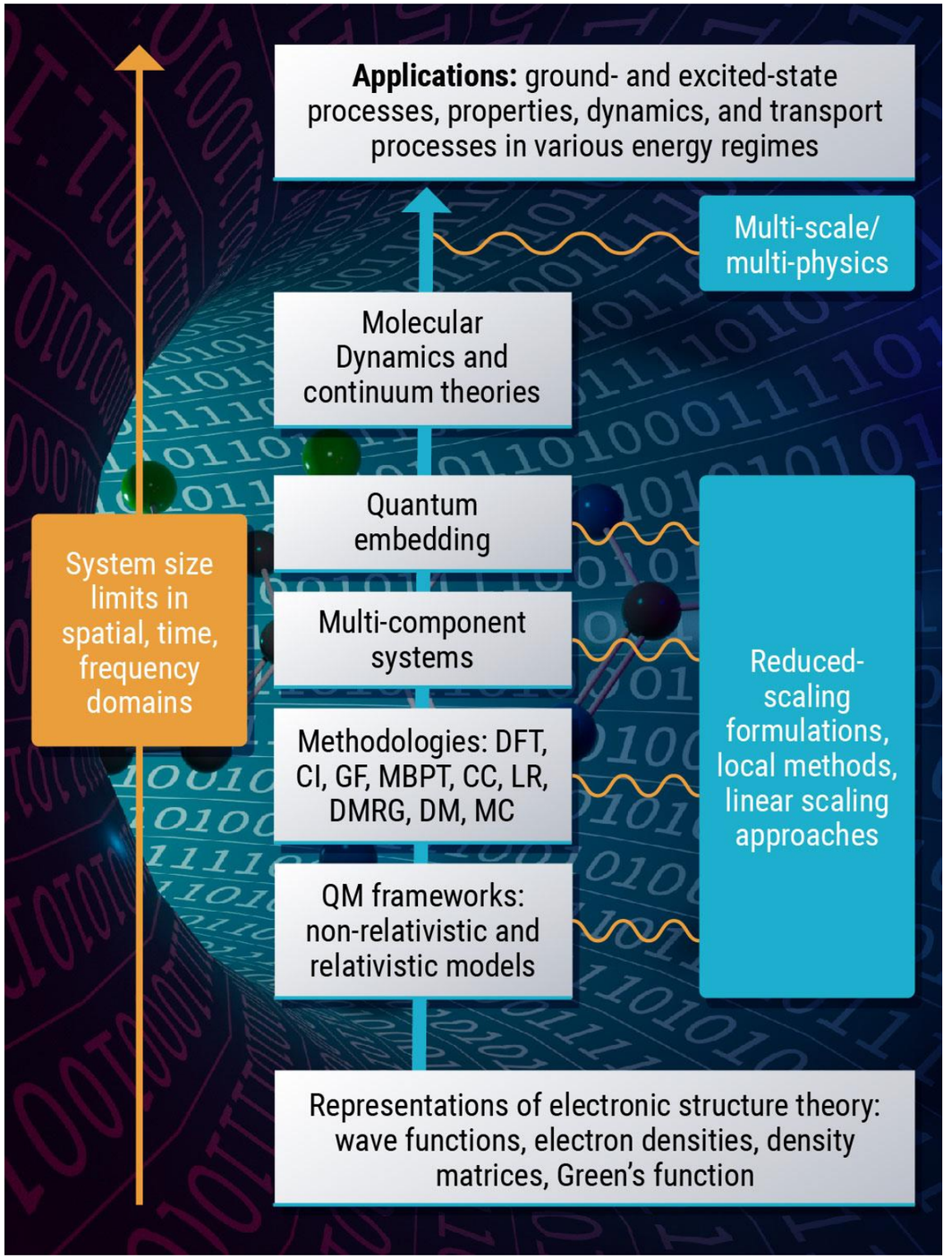
Details	
25	In-Person Attendees
16	Different Institutions
5	National Labs

Future of Software Development

- Community effort (continuous; update protocols as needed)
- Involve early career/underrepresented groups (MSIs)
- Stewardship
- Closely follow hardware development
- Address important science problems (scaling alone is not an end in itself)



Kowalski, Xantheas *et al*. "A Perspective on Sustainable Computational Chemistry Software Development and Integration, *J. Chem. Theor. Comput.* (under revision)



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Thank you