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DEPARTMENT: COMPUTER SIMULATIONS

Simulation for All: The Atomic, Molecular, and Optical Science Gateway

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The Atomic, Molecular, and Optical Sciences Gateway (AMOSGateway) enables novice and experienced users to utilize state-of-the-art software suites for tackling problems central to atomic, molecular, and optical science. This international collaboration provides a free platform and coordinated approach for computational research, allowing the community to produce new scientific results on an unprecedented scale. Currently hosting 10 software packages, the advanced cyberinfrastructure of AMOSGateway impacts many areas, including quantum information, cold atoms/molecules, plasma physics, and astrophysics. Future updates will further simplify these complex tools through graphical user interfaces, fundamentally transforming how practitioners gain expertise in cutting-edge computational research.

Computation has been an integral part of atomic, molecular, and optical physics since the development of the first computers. While the electromagnetic interactions governing the structure and dynamics of atoms and molecules are formally describable in a quantum mechanical framework, only the simplest cases are analytically tractable. Understanding electron correlation and how electrons are affected by external fields are central goals in the field of atomic, molecular, and optical science (AMOS). New insights can often only be gained through sophisticated simulations. Despite its long history of utilizing computation, AMOS is generally lacking a cohesive group effort for code sharing. Most codes are the property of individual research groups, prohibitively difficult to use, and rarely shared outside the group. Although some AMOS developers have released their codes to venues such as GitHub or Computer Physics Communications, building a viable executable on a local platform may become an insurmountable obstacle, as could accessing the supercomputer resources required to perform the calculations in a realistic amount of time.

The AMOSGateway[1] avoids that pitfall since users do not interact with either source code or supercomputing services directly but, instead, customize example input files and select supercomputing resources through a user-friendly interface on the AMOSGateway webpage. The gateway deals with the submission of job scripts, manipulates working directories, and carries out data retrieval once the calculation is complete. It even provides users with supercomputing resources via community allocations on national-tier machines.

Figure 1 shows schematically how the gateway interface serves as a hub to enable AMOS research and education. This methodology is a sea change for the AMOS community, opening the field to a larger and more diverse community of researchers by providing a vehicle to incorporate research-grade AMOS codes into university-level education and even making AMOS accessible to interested members of the general public.

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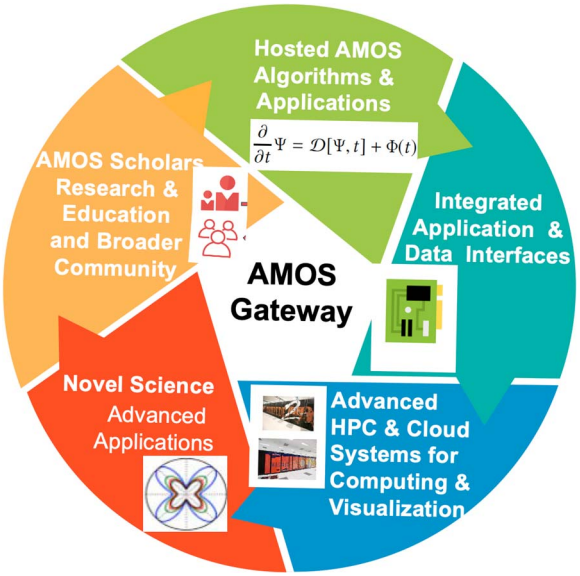

FIGURE 1. The AMOSGateway Cyberinfrastructure provides user-friendly interfaces and HPC resources, enabling scientific discovery as well as driving software development and education. AMOS: atomic, molecular, and optical science; HPC: high-performance computing.

The project was initiated during workshops held at the Institute for Theoretical Atomic and Molecular Physics at the Harvard Smithsonian Center for Astrophysics in 2018 and at the National Institute of Standards and Technology in 2019. Subsequently, the group obtained a modest start-up award from the National Science Foundation (NSF) through the XSEDE Extended Collaborative Support Services program, which provided the needed human expertise to deploy an existing set of state-of-the-art computer codes for general use by the AMOS community. In this article, we describe the capabilities of the gateway to enhance scientific productivity and provide an outlook to what the gateway developers plan for the near future.

SUPPORTED SOFTWARE SUITES

The principal purpose of AMOSGateway is to provide a broad set of users, who are interested in the dynamical properties of atoms and molecules and their interactions with external electromagnetic fields, with the computational tools required to achieve their goals, be they new science; the education and training of students; or, ideally, both. The currently hosted software suites fall into three main categories: structure codes, codes capable of describing steady-state collisions of electrons with atoms and molecules as well as first-order light–transition rates, and codes to treat the interaction of atoms and/or molecules with time-varying electromagnetic fields by solving the time-dependent Schrödinger equation (TDSE). A brief description of each software package as well as references for further information can be found at the AMOSGateway website.[1]

For atomic systems, widely used structure codes, such as the Dirac B-spline atomic R-matrix program (DBSR_HF)[2] and the general relativistic atomic structure package (GRASP),[3] are available on the gateway. They can be used in stand-alone mode for the calculation of energy levels and oscillator strengths. Furthermore, they can be employed to provide inputs to other applications that treat photon- and electron-driven processes, such as the B-spline atomic R-matrix (BSR)[4] approach and the convergent close-coupling (CCC)[5] method. The BSR approach is an alternative formulation of the well-known R-matrix method, capable of treating steady-state collisions with an (in principle) arbitrary, complex atomic or ionic target, while the CCC program can currently handle quasi-one- and quasi-two-electron targets. Both codes are fully parallelized using OpenMP and MPI, and CCC has already been ported to GPUs.

The gateway hosts three software suites capable of treating electron and photon collisions with general polyatomic molecular systems: UKRMol+,[6] MESA/Complex-Kohn,[7] and ePolyScat.[8] UKRMol+ computes low-energy electron and positron-scattering observables, photoionization, vibrational excitation cross sections for diatomic molecules, and permanent and transition dipole moments between bound and continuum states. MESA is a general polyatomic scattering suite employing the complex Kohn approach. It is particularly suited to describe processes with complex polyatomic systems, including interchannel coupling effects, dissociation, and extensive electron correlation via an ab-initio optical potential. ePolyScat computes electron–molecule scattering and molecular photoionization cross sections within the fixed-nuclei approximation, with the initial and final states described by a single-configuration state function. It is possible to extract rotationally and vibrationally resolved cross sections via the adiabatic nuclei approximation. Both UKRMol+ and ePolyScat are fully MPI-parallelized. An MPI version of MESA is currently under development.

The AMOSGateway hosts three additional software suites specifically focused on time-dependent phenomena: the R-matrix with time-dependence code (RMT),[9] time-dependent recursive indexing (tRecX),[10] and XUV/x-ray lasers for ultrafast electronic control in chemistry (XChem).[11] The RMT code has extended the R-matrix method to treat time-dependent processes in

atoms and molecules involving ultrashort, external electromagnetic fields, such as those occurring on the attosecond timescale. RMT's ability to describe electron correlation yields valuable insights that inform our understanding of light-mediated electronics in areas such as strong-field ionization, high-order harmonic generation, free-electron lasers, transient absorption, and streaking spectroscopies, which have shed light on the quantum phenomenon of tunneling.

The goal of tRecX is to establish a high-performance, flexible, well-documented, and robust code for solving the TDSE in the nonperturbative, strong-field, attosecond time-scale regime. Applications include the computation of fully differential single-electron emission spectra for atoms and molecules in the nonperturbative multiphoton and tunneling regimes for near-infrared radiation with elliptical polarization and arbitrary alignment of the molecular axis, high-order harmonics, Floquet spectra, and light-induced excitations and resonances.

XChem is capable of computing all-electron continuum wave functions of molecules by employing a hybrid Gaussian + B-spline basis, existing quantum chemistry packages for bound-state calculations, and close-coupling scattering methods. The code has been successfully used to describe the single-ionization continuum of atoms and small molecules to provide total and partial photoionization cross sections, resonance positions, and autoionization widths. Electronic and nuclear motion are treated on a level accessible only to few other codes. All three suites are parallelized via a combination of OpenMP and MPI.

To date, the complexity of these codes has limited their use almost exclusively to the developers and their research groups. The gateway removes these limitations and enables the latest versions to be deployed rapidly, along with the necessary support materials, in a way that will enable even nonexperts to exploit their sector-leading capabilities. This expanded user base also has the potential to include traditionally underrepresented groups in AMOS research, such as minority-serving and undergraduate institutions as well as researchers from outside the core fields of AMOS who are consumers of AMOS data. Plasma modelers and astrophysicists, who frequently use atomic and molecular collision data in their modeling, will be able to gain a deeper appreciation of how these data are generated by running the relevant codes themselves, thereby facilitating enhanced collaboration across subdisciplines of physics. The developers are dedicated to improving code documentation, to reducing the complexity of the workflows of the codes, and to lowering the entry barrier for new users. The high-performance computing (HPC) resources available on the AMOSGateway are provided to users without cost by the NSF.

AMOSGATEWAY CYBERINFRASTRUCTURE (CI)

Given the comprehensive nature of the gateway, an advanced CI is needed to provide the desired level of functionality. The AMOSGateway is built using the open-source Apache Airavata gateway middleware framework. The Science Gateway Platform (SciGaP)[12] is an NSF-supported project that serves multitenanted Apache Airavata middleware hosting services at Indiana University. It provides the servers and primary storage for the gateway. Airavata middleware also provides application registration, user-interface development environments, and file and other data management services. SciGaP delivers all of the services needed for a production gateway, such as gateway registration, maintenance, and an HPC registry. The middleware also manages the access to high-performance and cloud computational resources and scheduling systems. It provides job status updates as well as access to the output data and results in raw and basic postprocessed forms.

THE GATEWAY REMOVES THESE LIMITATIONS AND ENABLES THE LATEST VERSIONS TO BE DEPLOYED RAPIDLY, ALONG WITH THE NECESSARY SUPPORT MATERIALS, IN A WAY THAT WILL ENABLE EVEN NONEXPERTS TO EXPLOIT THEIR SECTOR-LEADING CAPABILITIES.

The basic functionality of the current gateway includes procedures to deploy applications and provides simple parameter definitions for each using an uploaded file or a set of files as a variable. Users can set job parameters on an HPC resource, including queue selection, wall time, and number of nodes/cores for a run. Postprocessing is currently limited to preexisting code execution as part of the submitted job. The Airavata workflow also includes task creation for scientific workflows, workflow information management, and user interfaces to the data needed for the workflows. This feature allows developers to create simplified workflows for their codes and novice users to easily execute multistep calculations. Users interact with the applications using a Django web front end, and they can access documentation and other resource pages created by the developers using the Wagtail content

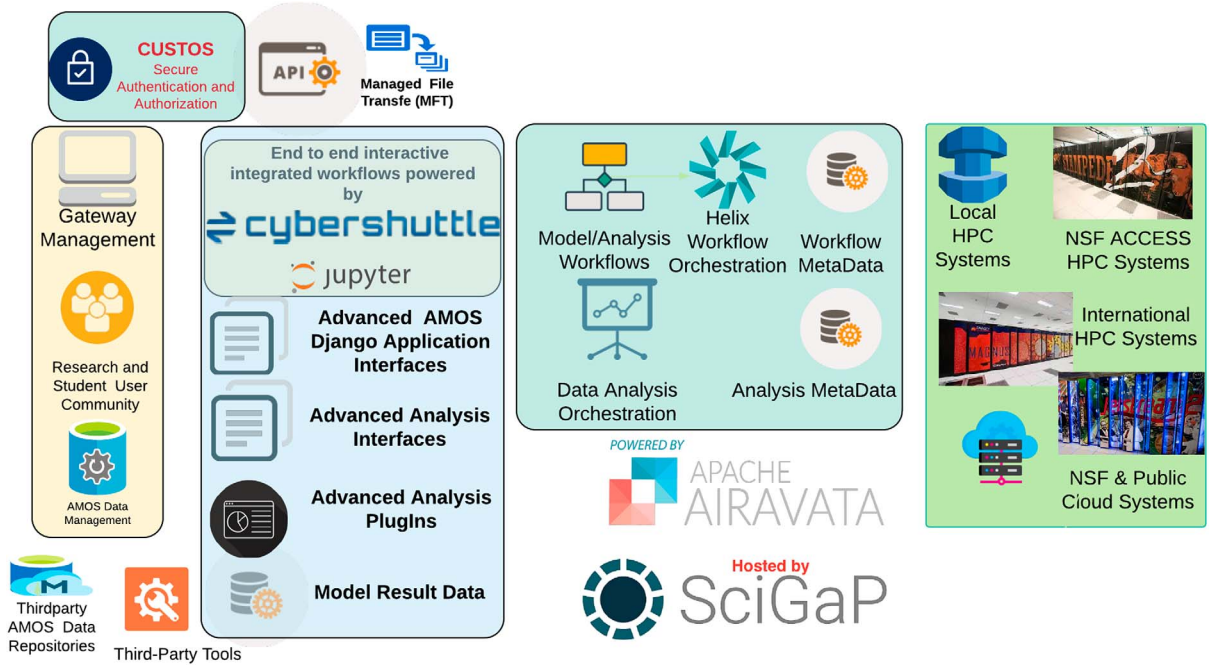

FIGURE 2. Proposed advanced AMOSGateway framework architecture integrating Apache Airavata middleware and Cybershuttle hosted by SciGaP services. API: application programming interface; NSF: National Science Foundation; SciGaP: Science Gateway Platform.

management system. The gateway computational workflows are processed using an Apache Helix workflow engine integrated into Airavata middleware. Some ACCESS resources, like Stampede2, Bridges2, and Expanse, are already integrated. User authentication is handled through the industry-standard Keycloak identity management system, thereby enabling most users to log in via their institution's single sign-on hub.

FUTURE DEVELOPMENTS

While the current version of the AMOSGateway has served as a resource for demonstrations and hands-on tutorials at several workshops, student training, and production-sized calculations, the recent receipt of NSF funding will enable the development of several new and upgraded features. New analysis plugins will be developed and integrated into the Django interface to enable the analysis of data obtained in complex, multistep experiments. Postcomputational analysis will be enhanced through the output metadata management system and mime-based third-party tool invocation available in the Airavata middleware as well as the new end-to-end research infrastructures to be developed by the recently funded NSF Cybershuttle project.[13] As shown in Figure 2, the Cybershuttle infrastructure will be integrated into the AMOSGateway to enable Jupyter notebook-based user interfaces and postprocessing developments.

New application interfaces will be developed using intuitive and flexible modules, with consistency checks and validation of input performed at each step. Workflows connecting multiple codes will also be developed. These workflows will be executed through the Apache Helix orchestrator integrated into Apache Airavata gateway middleware. An extensive series of user tutorials will be hosted on the gateway, accompanying regular webinars and workshops, along with sample educational materials detailing how instructors can incorporate gateway-hosted codes into their undergraduate or graduate classes. Finally, the creation of an AMOSGateway data portal will allow users to easily compare results obtained by different software suites.

CONCLUSION

AMOSGateway represents a comprehensive CI where AMOS practitioners can access a synergistic, full-scope platform for computational AMOS. Driven by a set of internationally recognized research groups in computational AMOS along with gateway software experts, the group is dedicated to making AMOSGateway the premier CI for researchers, students, and educators interested in AMOS. The codes hosted on the AMOSGateway have

produced some of the most cutting-edge computational research across the entire AMOS sector. AMOSGateway aims to make this level of computational research available to all—propelling scientific advances to be made not only by expert AMO scientists but by a larger, more diverse range of AMO scientists who previously have not had access to these resources.

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