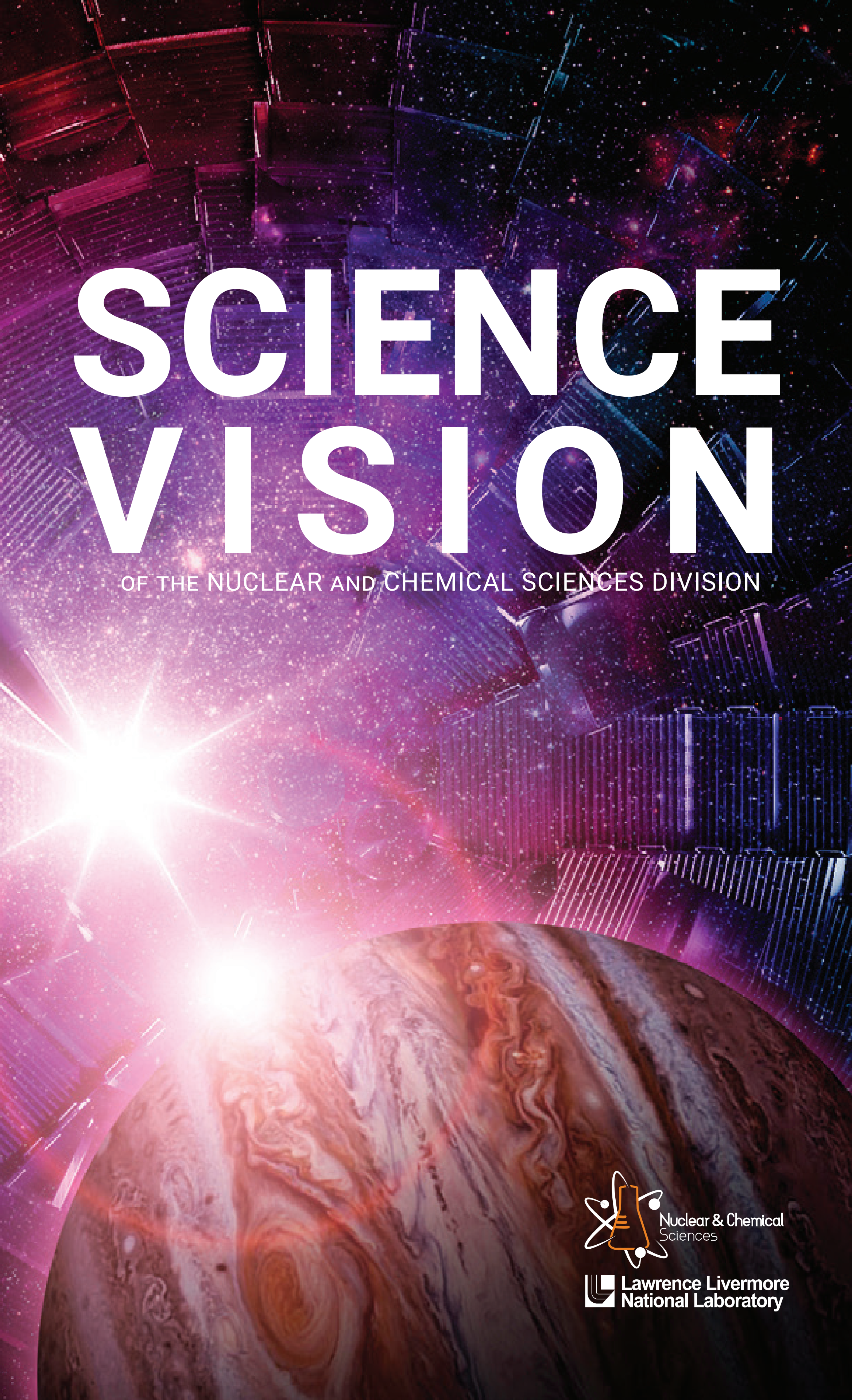
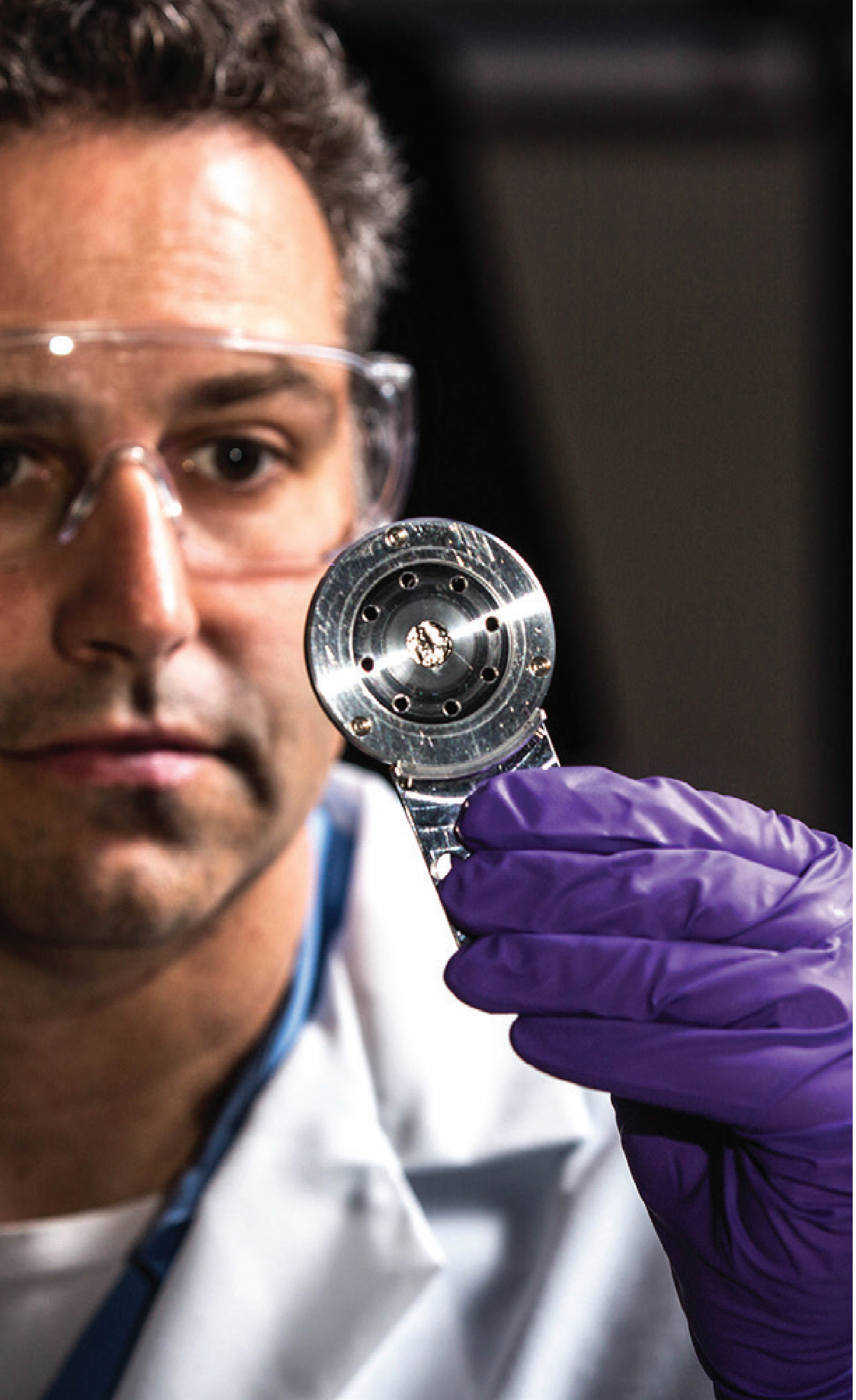


SCIENCE VISION

OF THE NUCLEAR AND CHEMICAL SCIENCES DIVISION

Nuclear & Chemical Sciences

Lawrence Livermore
National Laboratory



SCIENCE VISION

OF THE NUCLEAR AND CHEMICAL SCIENCES DIVISION

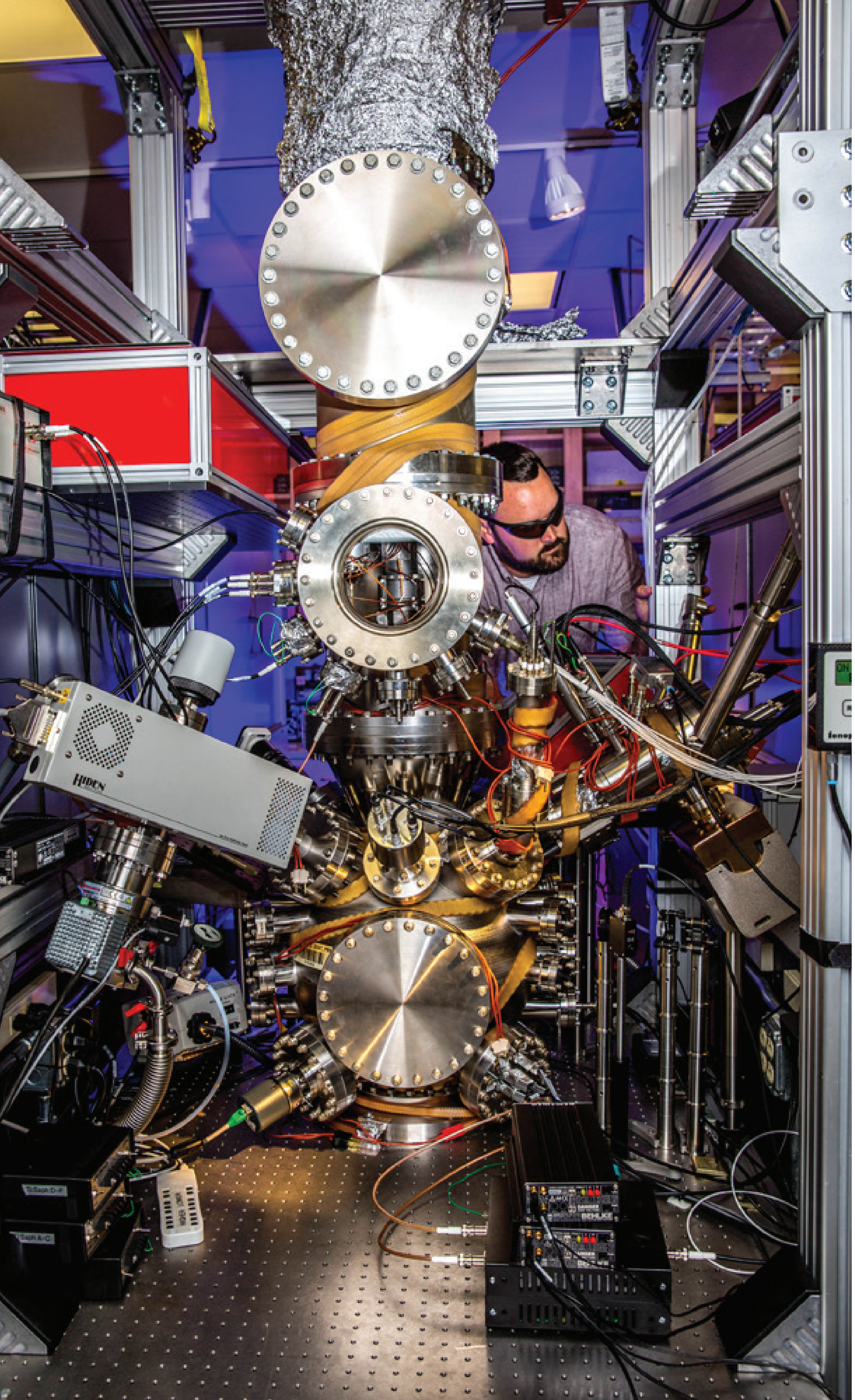
CONTENTS

> **“The history of science has proved that fundamental research is the lifeblood of individual progress and that ideas that lead to spectacular advances spring from it. ”**
>
> **— Sir Edward Appleton**
> *English Physicist, 1812–1965*

Our Mission

ON THE FRONTIER OF PARTICLE PHYSICS, NUCLEAR PHYSICS, AND CHEMISTRY

The Nuclear and Chemical Sciences Division advances scientific understanding, capabilities, and technologies in nuclear and particle physics, radiochemistry, forensic science, and isotopic signatures to support the science and national security missions of Lawrence Livermore National Laboratory.



Introduction

As a discipline organization at Lawrence Livermore National Laboratory (LLNL), the Nuclear and Chemical Sciences (NACS) Division provides scientific expertise in the nuclear, chemical, and isotopic sciences to ensure the success of the Laboratory's national security missions. The NACS Division contributes to the Laboratory's programs through its advanced scientific methods, capabilities, and expertise; thus, discovery science research constitutes the foundation on which we meet the challenges facing our national security programs. It also serves as a principal recruitment pipeline for the Laboratory and as our primary connection to the external scientific community.

This document outlines our vision and technical emphases on five key areas of research in the NACS Division—physics at the frontiers, structure and reactions of nuclei at the limits of stability, radiochemistry, analytical and forensic science, and nuclear detection technology and algorithms.

Physics at the Frontiers

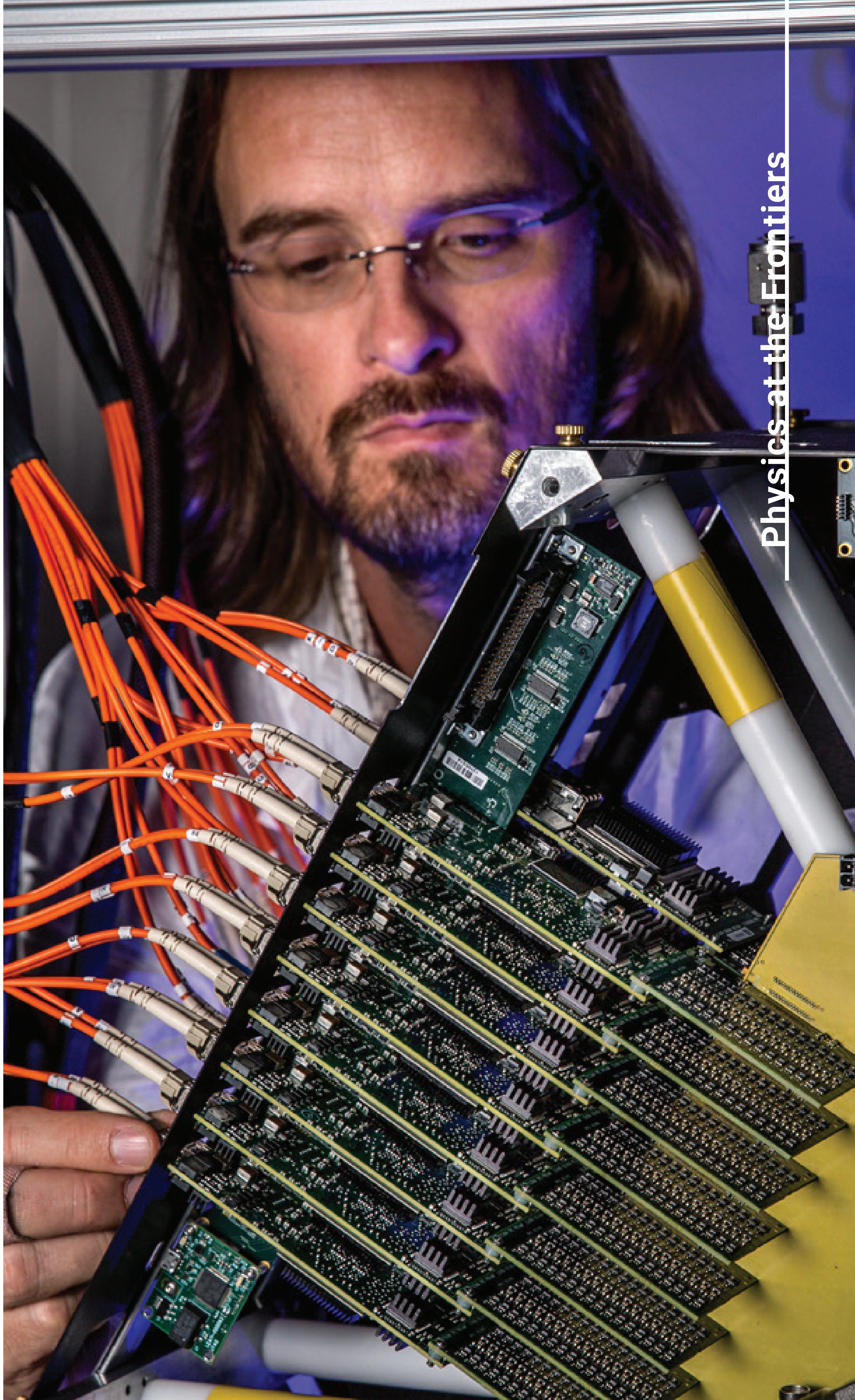
Understanding the fundamental forces of nature and the properties of the most elementary constituents of matter and energy drives research at the frontiers of modern physics. Only a small fraction of the universe is composed of the familiar baryonic matter (protons and neutrons). Indeed, about 25% of the mass of the universe is composed of an unknown dark matter that only interacts gravitationally, and about 70% is composed of dark energy, leaving only about 5% to ordinary baryonic matter. To further our understanding of the most fundamental constituents of nature and their interactions, NACS is pursuing compelling scientific problems through collaborations on key Department of Energy (DOE) Office of Science Projects.

Physics Beyond the Standard Model

The discovery of the Higgs boson has confirmed much of our understanding of the nature of the electroweak interaction, as embodied in the Standard Model of Weinberg, Salam, and Glashow. Many questions, however, remain unanswered and the Standard Model is universally thought to be incomplete. Further experiments are needed to search for physics beyond the Standard Model to define the fundamental forces of nature.

For example, the neutrino is among the most extraordinary of the elementary particles. Since its discovery in 1959, key questions remain: (1) Are there more than three types of neutrinos (including a possible sterile neutrino)? (2) What are the neutrino masses? (3) Are neutrinos their own antiparticles? (4) Do neutrinos violate charge-parity symmetry? Our unique combination of capabilities, expertise, and relationships position us to lead or collaborate in flagship experiments addressing these key questions.

The Standard Model predicts three massless flavors of neutrinos associated with each leptonic flavor. This picture is incomplete, as experiments over the past several decades have demonstrated that neutrinos must have mass by exhibiting oscillations between the flavors. In addition, there is the tantalizing possibility that neutrinos may be their own antiparticles and the



possibility of a fourth, so-called sterile neutrino. LLNL is a founding member of the PROSPECT (Precision Oscillation and Spectrum Experiment) collaboration, and the lead laboratory for the deployment and operation of the PROSPECT detector at the Oak Ridge High Flux Reactor. This experiment seeks to determine whether a new type of neutrino exists and to measure the flux of reactor antineutrinos with unprecedented accuracy. Even if a new neutrino type is not discovered, PROSPECT will have developed a novel antineutrino detection technique that permits above-ground operation of near-field (less than ~500-meter standoff) detectors, as well as improved spectral resolution for reactor antineutrinos. These advances are essential to broadening the applicability of antineutrino-based reactor monitoring in nonproliferation and safeguards contexts—a field pioneered and developed by LLNL scientists.

LLNL is also the founder and lead institution for the WATer CHerenkov Monitor for AntiNeutrinos (WATCHMAN). WATCHMAN is an underground, 1000-ton water Cherenkov detector, doped with the neutron-capture agent gadolinium. The doping technique, invented by LLNL scientists in 2001, permits high sensitivity to the mega-electronvolt-scale antineutrinos emitted from reactors, using a low cost and scalable medium. By observing reactor operational transitions at 25-kilometer standoff, WATCHMAN will demonstrate for the first time that water detectors, when doped with gadolinium, can be made sensitive to reactor antineutrinos. This capability has relevance for reactor monitoring regimes, such as the International Atomic Energy Agency's Safeguards regime. WATCHMAN is also integral to the DOE Office of Science long-range plan for neutrino detection. Water Cherenkov technology, built at the megaton scale, is one of only two large-scale detection options for the Long Baseline Neutrino Facility (LBNF)—the U.S.'s flagship experiment for the coming decade. WATCHMAN will serve as a large-scale test-bed to develop and study particle reconstruction, background rejection, light collection, and other techniques that will be needed for the still larger scale LBNF water Cherenkov detectors. In addition to its reactor monitoring and technology development applications, WATCHMAN will be one of the world's largest supernova detectors, with a unique ability to distinguish between the neutrino and antineutrino components of the supernova burst and an unprecedented pointing accuracy. This pointing capability is important since the neutrino burst precedes the optical signal from supernovae by several hours, providing for the more accurate cueing of optical telescopes to be trained on the supernova as quickly as possible.

Double-beta decay, in which two electrons and two neutrinos are emitted simultaneously is an extremely rare process predicted by the Standard Model and known to occur in a small number of nuclei. More significant is the possibility of double-beta decay in which no neutrinos are emitted—so-called neutrinoless double-beta decay, or $0\nu\beta\beta$. If observed, this decay would be a direct signal for physics beyond the Standard Model, as it is allowed only if neutrinos have mass and exist as their own antiparticles. Observation of $0\nu\beta\beta$ could provide clues to the origin of mass and the preponderance of matter over antimatter in the universe. Current limits indicate that the lifetime for this decay mode must be greater than 10^{25} years. To detect this decay mode, large amounts of specially processed material are required within a carefully controlled background environment. Unique expertise in time-projection chamber technology has enabled LLNL to assume a leading role in the nEXO (next Enriched Xenon Observatory) collaboration, which plans to field a ton-scale time-projection chamber detector wherein xenon-136 serves both as the decay source and detector. Over the next few years, NACS will lead the research and development effort for nEXO with the goal of winning the competition for the down-selection to field a U.S.-led experiment. Presently, all the proposed $0\nu\beta\beta$ experiments have a sensitivity of detection only in the case where the electron neutrino is heaviest of the three types of neutrinos, or in what is referred to as the "inverted hierarchy." NACS will drive the development of science capabilities for nEXO while also engaging in technologies for next-generation experiments with the sensitivity to detect lighter neutrinos in the "normal" hierarchy.

The $0\nu\beta\beta$ experiment is indirectly sensitive to the mass of the electron neutrino, which may be as low as a few milli-electronvolts. A direct experiment infers the neutrino mass from the missing energy in the endpoint of the beta spectrum in tritium beta decay, which, thus far, has provided a limit of approximately 1 electronvolt. These experiments are complicated by atomic and molecular effects that are difficult to control or accurately predict. LLNL has joined the Project 8 collaboration and is providing expertise on tritium molecular recombination to greatly increase the experimental reach of tritium endpoint experiments toward a measurement of the neutrino mass.

While the broad physics program at the European Organization for Nuclear Research's (CERN's) Large Hadron Collider (LHC; Geneva, Switzerland) has been a stunning success with the discovery of the Higgs boson and by placing severe limits on many favored new physics models, such as supersymmetry and technicolor, evidence for any physics beyond the Standard Model has remained elusive. As the LHC pushes into the extreme range of its energy and luminosity performance, the challenge becomes even greater to extract hints of new physics from immense backgrounds. For more than a decade, LLNL has helped pioneer new and innovative techniques to enhance the discovery potential of collider experiments, such as identifying events in which beam protons remain intact after undergoing a hard scatter via photon or gluon exchange, which produces a high-mass central state that decays into other particles. By tagging on outgoing protons, we can select particles from this high-mass central state that produce electroweak bosons, which are particularly sensitive to nearly all new physics models, especially those related to the Higgs boson. These tagged interactions provide a model-independent means to detect the presence of new particles in a theoretically clean manner.

NACS currently leads the construction and commissioning of the picosecond clock system, essential for combining the proton-tagging information from the detector stations. We are also leading the physics analysis of the exclusive production of W-boson pairs, which is the most promising channel for discovering new physics from proton tagging. By extending the analysis to include proton tagging, we hope to achieve the sensitivity required for new and revolutionary physics discoveries with LHC data.

Physics of the Big Bang

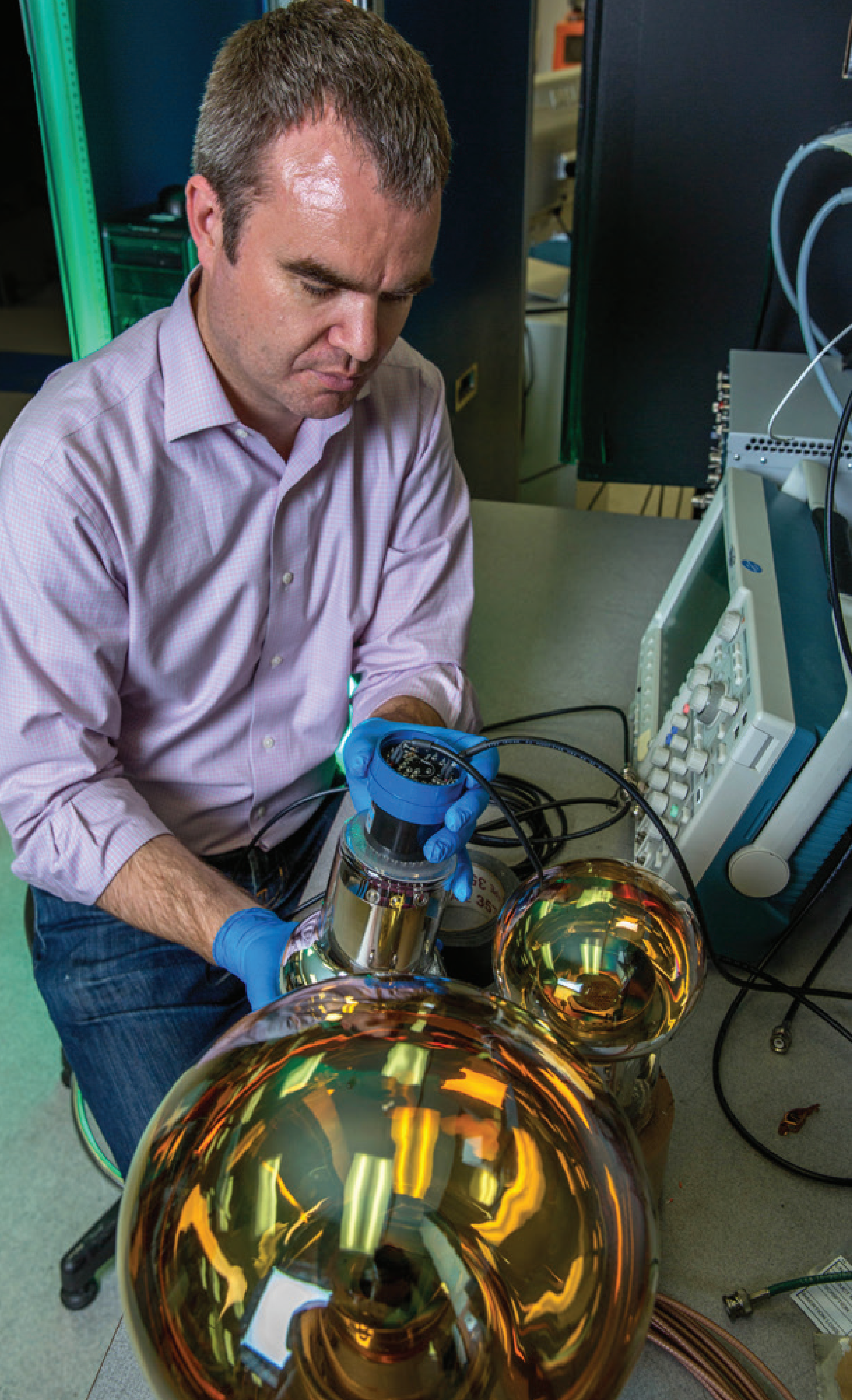
Ultra-high-energy collisions between nuclei at Brookhaven National Laboratory's Relativistic Heavy Ion Collider (RHIC; Upton, New York), and at the LHC, create the conditions needed to reveal emergent phenomena present when the universe was approximately 1 microsecond old and at a temperature of 10 trillion Kelvin. Research with relativistic heavy ions has been richly rewarded with the discovery of a strongly interacting plasma of quarks and gluons (quark–gluon plasma, or QGP) with a viscosity at, or near, the quantum limit for liquids and conditions exhibiting a duality with strong gravitational fields surrounding black holes. Nonetheless, pressing questions remain regarding the substructure of the QGP and the existence of critical behavior in the phase diagram. Upgrades and experiments are now underway to address these questions as this field prepares for the Electron Ion Collider (EIC), a key tool in understanding the gluonic matter that dominates atomic nuclei at short distances (high energies).

LLNL has made significant contributions to both hardware (PHENIX [Pioneering High Energy Nuclear Interaction Experiment] magnet design and Tier-2 LHC Grid Computing) and physics analysis, including space–time measurements to constrain models of the QGP evolution at RHIC and lattice quantum chromodynamics (QCD) calculations of the equation of state and transition temperature using LLNL's supercomputers. These scientific interests have led naturally to comprehensive models of heavy-ion collisions and measurements that will produce the best constraints on the initial conditions and temporal evolution of plasma. Early work at LLNL led to sophisticated model-to-data comparisons that now constrain the hydrodynamic evolution of the QGP, and we are now working within the sPHENIX experiment and JETSCAPE (Jet Energy-loss Tomography with a Statistically and Advanced Program Envelope) collaborations to further develop these methods and elucidate the substructure of the QGP.

Highly energetic jets of particles emitted from the QGP offer a sensitive tool to probe detailed properties of the QGP, providing information about the substructure of the plasma, evolution and transport of quarks and gluons in the plasma, physics prior to the formation of the equilibrated plasma, and the correspondence between anti-de Sitter spaces and conformal field theories. NACS research focuses on implementing Bayesian methods to explore new methods for finding and analyzing jet probes and comparing them to model predictions. As the understanding of jets in heavy-ion collisions becomes more sophisticated, our goal is to elucidate the methods most connected to the underlying physics. This can be achieved through simulations for sPHENIX and the analysis program of the JETSCAPE collaboration. In addition to advancing our understanding of the QGP, our heavy-ion work will continue to serve as a magnet for recruiting and developing skills in complex data analyses, modeling and simulations, scientific computing, and novel detector building.

Theory of Quantum Chromodynamics

Quarks are bound into mesons and hadrons by QCD—a force so strong that quarks cannot be isolated and studied individually, and only the most powerful supercomputers can compute their properties. QCD not only defines the strong force that binds quarks within mesons and baryons, but also the force between nucleons in the nucleus. Although nuclear theorists model the effect of the strong force with potentials, the correct underlying physics is QCD. However, the extremely strong nature of QCD and the fact that gluons, the force mediator, are self-interacting make QCD nonperturbative and intractable to many of the traditional methods in theoretical physics. QCD must be solved using a path-integral formulation on a discretized space–time lattice with the aid of high-performance computing. LLNL is a leader in these lattice QCD computations, which promise to answer many fundamental questions about the properties of elementary particles and how they interact. Within the framework of lattice QCD, we are focusing on understanding the interplay between quarks and the weak interaction, such as the nucleon weak axial charge. Lattice QCD calculations afford a unique insight into the interaction between nucleons in a nucleus that will guide nuclear theorists aiming at *ab initio* descriptions of nuclei. We will also use lattice QCD calculations to analyze potential mediators of $0\nu\beta\beta$ and calculate the requisite matrix elements needed to properly interpret and analyze experiments in the event $0\nu\beta\beta$ is observed.



Detecting the Elusive Dark Matter

In dark matter physics, LLNL is a founding member of the XENON-10, Large Underground Xenon (LUX), and LZ collaborations, all of which have sought or will seek to directly detect interactions between dark matter particles and normal matter here on Earth. These detectors have enjoyed a Moore's law level of improvement of target mass and sensitivity, with deployed experiments increasing from the 10-kilogram size in 2004 (XENON-10) to the 300-kilogram size in 2010 (LUX). The LZ experiment will provide approximately another tenfold increase in mass and a hundredfold increase in sensitivity, which is nearing the sensitivity limit for these types of detectors that is determined by the confounding and irreducible background arising from atmospherically generated neutrinos. NACS specializes in studying the properties of dual-phase detectors at their limits, including the lowest-ever measurement of the ionization yield in argon. NACS research will determine the ultimate sensitivity of liquid noble gas detectors to dark matter interactions, and will explore the possibility of a new, high-rate method for detection of reactor and solar neutrinos by exploiting coherent neutrino-nucleus scattering.

LLNL is also a founding member of the Axion Dark Matter Experiment (ADMX), and is heavily involved in both leadership and technical roles. This project is the flagship U.S. search effort for dark matter axions, a well-motivated alternative to weakly interactive massive particles that is highly complementary to the LUX/LZ dark matter search. It is the only experiment that has probed the plausible axion dark matter phase space and is poised to have high discovery potential over the next few years as it begins operating with unprecedented sensitivity. Detection of dark matter axions would answer several fundamental mysteries in physics and cosmology. NACS will lead the microwave cavity working group with responsibility to deliver higher frequency resonator systems, which includes microwave cavity and superconducting detector development—a technical role synergistic with recent efforts in superconducting quantum sensors.

Structure and Reactions of Nuclei at the Limits of Stability

Studying atomic nuclei is essential to our understanding of the evolution of the universe, as nuclei comprise more than 99% of the mass of the visible matter in the cosmos. Nuclear reactions not only power stars, but they are also responsible for the synthesis of all the elements found on Earth. Research in the fundamental properties of atomic nuclei is entering an exciting new era that promises to shed light on many key questions in nuclear physics. This renaissance is driven by two factors: (1) new facilities, such as the Facility for Rare-Ion Beams (FRIB) at Michigan State University (MSU), which offers an unprecedented opportunity to study nuclear properties, especially neutron-rich nuclei near the limits of stability; and (2) high-performance computing, which, for the first time, is allowing theoretical descriptions of complex nuclei from first principles. Our vision for research in low-energy nuclear physics will push the frontiers of understanding by weaving together NACS's capabilities in theory and experiment to fully capitalize on our nation's investments in FRIB and high-performance computing.

Exploring the Limits of Nuclear Stability

The U.S. nuclear physics community has reaffirmed that the path to understanding atomic nuclei requires FRIB. This new facility will provide unprecedented access to new and exotic nuclei that will lead to scientific breakthroughs and significant advancements in our understanding of nuclei and their role in the cosmos. In particular, FRIB will enable investigations of neutron-rich nuclei, which is currently the most fertile ground for nuclear structure research. This research will help determine the nature of the strong force between nucleons, how nuclei are constructed, and how complex systems display regular patterns and symmetries. By probing the limits of existence near the neutron-drip line, FRIB will explore how nucleosynthesis formed the elements by successive neutron capture during the r-process. Ultimately, FRIB will provide the experimental data required to lay the foundation for a unified, predictive

theory describing how nuclei are put together and how they interact.

The r-process proceeds through successive neutron captures involving many short-lived, neutron-rich isotopes far from stability. Understanding the r-process requires significant new data, such as beta-delayed neutron emission probabilities along the decay sequence back to stability, neutron-capture cross-sections for key nuclei, and a detailed understanding of the evolution of shell structure in nuclei.

When coupled with the Gamma-Ray Energy Tracking Array (GRETA), the LLNL-designed CHICOx (the latest version of the Compact Heavy Ion Counter) detector system will provide an ideal laboratory for studying the structure of r-process nuclei through quasi-elastic reactions. Pioneering experiments performed by NACS with reaccelerated CARIBU (Californium Rare Isotope Breeder Upgrade) ion beams delivered to CHICO2 (coupled with GRETINA, an in-beam gamma-ray array) have already demonstrated the sensitivity of this approach by revealing the octupole nature of barium-144, a short-lived, neutron-rich isotope (half-life of 11.2 seconds). At FRIB, this research program will provide valuable information about how nuclear properties such as correlations, collectivity, and configuration coexistence evolve as nuclei move further away from stability and approach the neutron drip line.

An LLNL-led team has established a powerful new approach to studying beta-delayed neutron emission using the beta-decay Paul trap. With the increased intensity of low-energy beams from FRIB, this approach can be used to determine the beta-delayed neutron emission properties of isotopes extending to neutron-rich nuclei along the r-process path. The addition of neutron-detection capabilities will permit multiple neutron-emission branches to be studied.

The telltale signatures of physics beyond the Standard Model can be revealed by searching for deviations from the predictions of electroweak theory in nuclear beta decay or in the discovery of an electric dipole moment (EDM) in nuclei. With ion traps, the nuclear recoil following beta decay, as well as the neutrino momentum and all decay correlations, can be determined. Recently, a high-precision measurement of the beta-neutrino angular correlation from the decay of lithium-8 was carried out using the beta-decay Paul trap. This resulted in a more stringent limit on a broad class of additional contributions to the electroweak interaction. Coupled with ion traps and new detector systems, the high-intensity beams at FRIB will allow improved tests of the symmetries that govern the interplay between the weak, electromagnetic, and strong nuclear forces.

An EDM signature can be amplified by several orders of magnitude in nuclei such as radium-225, radon-223, or protactinium-229, which are expected to have a pear-like, or octupole-deformed, shape. Octupole-deformed nuclei are promising candidates for EDM searches; however, the isotopes anticipated to have the largest sensitivity are typically heavy, short-lived isotopes previously inaccessible to detailed study. A program to quantify the degree of octupole deformation in exotic nuclei can be performed with FRIB beams delivered to CHICOx and GRETA. After which, LLNL radiochemistry expertise can be leveraged to efficiently harvest these isotopes from FRIB and prepare them for an EDM search.

Neutron-capture cross-sections for key short-lived isotopes can be determined using indirect methods that populate the excited compound nucleus through "surrogate" reactions in inverse kinematics or nuclear beta decays, which can provide valuable information from which modelers can infer the reaction of interest. NACS has significant expertise in surrogate reactions and beta decay, and the high-intensity beams from FRIB, combined with a robust theoretical description of reactions, will pave the way toward the determination of cross-sections for isotopes produced with intensities as low as 1 ion per second.

In addition to FRIB, dedicated photon and neutron sources located in the NACS accelerator complex will provide LLNL researchers with capabilities to perform precise experiments for photon- and neutron-induced reactions on nuclei. A recently acquired, state-of-the-art 2.998-gigahertz electron accelerator with a thermionic source will operate as a high-flux bremsstrahlung source. This capability will permit precision measurements of the gamma strength function in nuclei, photo-fission processes, and nuclear resonance fluorescence. A dedicated neutron source will also be available via a high-current deuteron-ion accelerator with a deuterium gas target. Coupled with a hot-cell to handle radioactive materials, this facility could ultimately make direct measurements of neutron-induced reactions on short-lived nuclei for nuclear data and astrophysics applications that cannot be performed elsewhere.

Predicting Nuclear Properties

Over the past decade, the LLNL nuclear theory group has helped pioneer *ab initio* descriptions of atomic nuclei, which formulate the beginnings of a unified theory for nuclei. This effort was advanced with multiple investments and ample access to LLNL supercomputing capabilities. Our research has focused on calculations for light nuclei utilizing realistic forces between the constituent nucleons within a microscopic framework. Early work demonstrated the impact of the three-nucleon force on the low-lying structure of nuclei. This theoretical framework has now been extended to a full theory, and NACS is leading a national effort in the *ab initio* description of light-ion reactions, such as d(t,n)^{4}He. More complex reactions, such as ^{3}He(^{4}He,γ)^{7}Be, will soon be viable. *Ab initio* methods are now approaching the neutron drip line and exposing weaknesses in our understanding of the nuclear force. Our efforts continue to advance this research with a focus on *ab initio* descriptions of reactions, the structure of drip-line nuclei, and collective excitations in light nuclei, and in seeking full analytical understanding of the fundamental interactions between nucleons.

To understand the structure of heavier nuclei, LLNL has joined the SciDAC-3 (Scientific Discovery through Advanced Computing) collaboration to develop a more robust framework based on nuclear density functional theory methods. The goal is a formalism to enable predictive calculations for heavy nuclei, such as super-heavy elements, fission, and simulations of nucleosynthesis during the r-process. Much like truly *ab initio* approaches, the objective is to develop a coherent density functional theory based on nuclear forces guided by chiral effective field theory.

A full theory for nuclei goes beyond answering the question of how nuclei are constructed to accurately describing how they interact. Today, reaction theory is overly reliant on highly approximate models determined empirically for specific regions in the nuclear chart. Such models lack the predictive capability to describe systems beyond their empirical formulation and have unquantified uncertainties. These empirical models are a primary limitation of the usefulness of the "surrogate" method. Consequently, a generalized, predictive theory for nuclear reactions is required. Inspired by the success of the *ab initio* program for light nuclei, we are pursuing a hybrid approach to nuclear reactions for heavy nuclei that combines density functional theory to depict the target-plus-projectile system with microscopic R-matrix theory to describe the reaction dynamics.

Theoretical calculations are often based on input parameters, such as the those defining the nuclear force, that are empirically constrained with experimental data. The ability of these parameters to reproduce these data defines their uncertainties. We are exploring approaches utilizing Bayesian methods and machine learning techniques to quantify how these uncertainties, as well as those induced by theoretical models, affect our theoretical predictions for nuclear properties and their impact on applications such as nucleosynthesis.

Radiochemistry

Research in radiochemistry explores nuclear reactions, the limits of nuclear stability, and the properties of the heaviest elements. The development of new chemical separation and automation methods, diagnostics, and experimental platforms constitutes the foundation for solving challenging problems, such as the production and isolation of unique isotopes to enable nuclear reaction studies, the exploration of nuclear reactions in a plasma, and, ultimately, the study of chemical properties of single-atom species. The fundamental nuclear science covered by these studies ranges from light-particle capture to one-atom-at-a-time heavy-element chemistry and, therefore, requires faster, more precise, and more effective technical approaches to push the boundaries of the periodic table and our understanding of chemical properties.

One area of focus is the study of how radiochemical processes are affected by plasma environments, such as those found in a thermonuclear detonation or the burning interior of a star. Fractionation of chemical elements and isotopes occurs over a broad range of spatial and temporal scales, from the accretion of solar systems to the phase separation of molecules. Although nuclear explosions, by recreating stars on Earth, offered a window into isotope formation, the scientific information obtained in these events was limited by condensation, fractionation processes, and the rapid timescale and extreme conditions in which these events took place. We are developing and testing experimental techniques that allow us to identify the chemical species present during the rapid condensation of the plasma and to constrain the conditions under which they form. Two experimental methods are being used: a unique inductively coupled plasma-flow reactor and a laser-induced breakdown spectroscopy apparatus that enables redox-controlled experiments. Diagnostics include *in situ* spectroscopic measurements and *ex situ* characterization of particle chemistry. Our long-term goal is to use these tools to test assumptions in nuclear fallout models and validate chemical kinetics and microphysics codes.

Recreating Stars on Earth

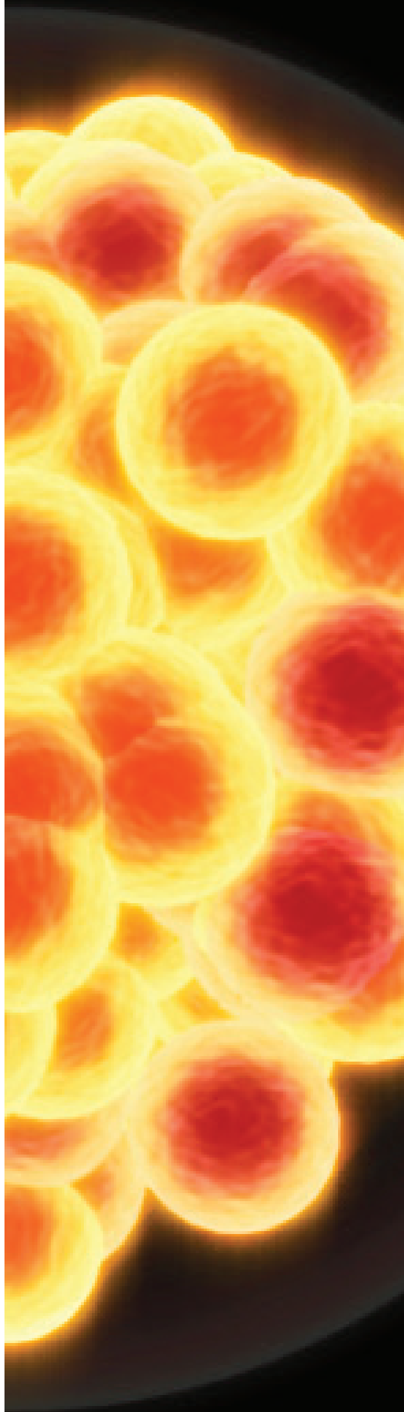
Livermore's own National Ignition Facility (NIF), which creates temperature and density conditions mimicking the interior of a star, offers a unique environment to study the effects of plasma on condensation and nuclear reaction processes. During a typical NIF shot, 192 lasers impinge on a 2-millimeter-diameter capsule filled with equimolar deuterium–tritium (DT) gas. The capsule is compressed, which causes the deuterium and tritium nuclei to fuse, forming a large flux of 14.1 mega-electronvolt neutrons. In the process, the capsule target assembly is vaporized in a plasma environment, and the resultant material is collected by solid and gaseous debris radiochemistry collectors. Patterns in the collected debris indicate strong effects of the plasma on the subsequent condensation of the debris, and the fractionation of debris products has been observed. The processes that govern these fractionation effects have been shown to be a mixture of differing condensation rates during plasma cooling as well as differences in chemical bonding between elements. We also study the process of condensation at NIF using depleted uranium hohlraums and capsules, which produce fission products when they interact with the neutrons emitted from DT fusion. Using different materials and methods for debris collection, we can begin to understand both the chemical and mechanical processes that dictate the behavior of elements cooling from a plasma environment.

NIF also offers the unique opportunity to study the effects of a plasma environment on nuclear reaction kinetics. During a typical accelerator irradiation, the neutral target atoms create an electron screen for the incoming projectile nuclei, lowering the Coulomb potential between the nuclei and resulting in a higher reaction rate. On the other hand, for nuclear reactions occurring in a stellar interior, the nuclei are ionized and, therefore, do not suffer from the same bound-electron screening that is present during an accelerator experiment. The plasma environment results in a higher Coulomb potential between the reactant nuclei, thus lowering the reaction rate. Therefore, accelerator measurements cannot be extrapolated to stellar conditions without models that attempt to describe the effects of bound-electron screening. Measuring nuclear reactions from the inside of a NIF capsule would more closely resemble the environment found in a stellar interior in a high-energy-density plasma, and models of the production of nuclei in the cosmos ultimately depend on having accurate data to inform them.

Measuring nuclear reactions in a NIF-generated plasma requires the development of unique platforms and diagnostics. First, we are studying mechanisms to add dopant target atoms into the NIF capsule shell. We have created a capsule injection station where small numbers of atoms (~10^{16}) can be added to the inner surface of a NIF capsule shell through the fill-tube hole in a solvent, which is then dried, leaving behind a single uniform layer of target atoms on the inner surface of the shell. The number of atoms has been shown in previous NIF shots to be large enough to produce a measurable number of nuclear reactions and not adversely affect the fusion-neutron yield of the capsule. Continued research will establish methods for adding radioactive species to NIF capsules and enabling measurements of nuclear reactions on short-lived species in a plasma environment, which cannot currently be achieved at traditional accelerators. In addition, we continue to develop new diagnostics for improving the collection of solid and gaseous debris from NIF shots. Among our goals is to study reactions relevant to stellar nucleosynthesis in a true plasma environment, which can then be compared with model calculations to improve our understanding of how plasmas affect nuclear-reaction rates. As fusion yields increase, a unique potential arises for measuring nuclear cross-sections on excited nuclear states. The burn time of a typical NIF DT capsule is much shorter (tens of picoseconds) than the lifetime of some nuclear excited states. By loading a capsule with appropriate radionuclides, the contributions to the overall cross-section from excited states could be deduced. Such experiments would ultimately require higher neutron yields from NIF fusion capsules, and we are establishing the platforms and diagnostics needed to perform these measurements.

Exploring the Properties of Short-Lived and Heavy Elements

New methods and techniques for radiochemical separations are being developed at NACS to speed up processes for isolating radioactive species. Some of these methods include the development of chemical automation processes in which a solution of radionuclides comprised of several elements is separated into its individual components with time scales ranging from a few seconds to a few minutes. This type of automated separation can apply to isotope harvesting, where unique isotopes produced as byproducts in accelerator irradiations are collected and used in radiochemistry experiments. For example, work is under way at the National Superconducting Cyclotron Laboratory (NSCL) at MSU in preparation for future experiments at FRIB. In collaboration with Hope College, the University of Alabama, and the University of Notre Dame, a new beam-stop has been installed at NSCL where radionuclides produced from irradiation can now be collected in a liquid coolant circulation line, which can then be processed radiochemically for removal of specific isotopes. As these are high-radioactivity samples, remote chemical automation must be investigated to process materials for subsequent handling and study. Our initial experiments



performed at MSU show promising results for isotope harvesting at this new beam-stop, and separation methods specific to collecting zirconium nuclides have been successfully tested. Automation and the flexibility to "dial" any element of interest into the system will enable FRIB to continually harvest nuclides for nuclear medicine, chemistry, and physics.

Chemical automation is also critical for studying the chemical properties of the heaviest elements. Elements with atomic numbers greater than 104 are created with extremely low production rates—on the order of 1 atom per day or less. Therefore, studying their chemical properties requires a chemical system that is specific to the element of interest; has a high-separation, high-selectivity factor from interfering activities; and is extremely rapid so that the system can enter pseudo-equilibrium in 1 second. The chemistry of lighter transactinides is known, and current research is focusing on elements with atomic numbers greater than 112, especially element 114 (flerovium), which is key due to conflicting models of its atomic properties and how they might be affected by relativistic effects from the massive nuclear charge. Based on its chemical group alone, flerovium would be expected to chemically behave like lead, but models predict a behavior more like mercury or radon; the latter would make flerovium the next noble gas. Understanding flerovium's behavior would allow its proper placement in the periodic table based on its chemical properties rather than atomic number. The longest-lived isotope of flerovium has a half-life of 2 seconds, which means chemical separation must occur within 1 second. Specific ligands designed to pull flerovium from a solution based on its ionic radius are currently being designed and synthesized for study of the homolog elements of group 14. Once the chemical system has been established, similar procedures will be used for the separation of flerovium to determine its behavior compared to neighboring elements in the periodic table. These experiments will be conducted at the GSI Helmholtz Centre for Heavy Ion Research (Darmstadt, Germany) and would constitute the first reported aqueous chemistry of element 114.

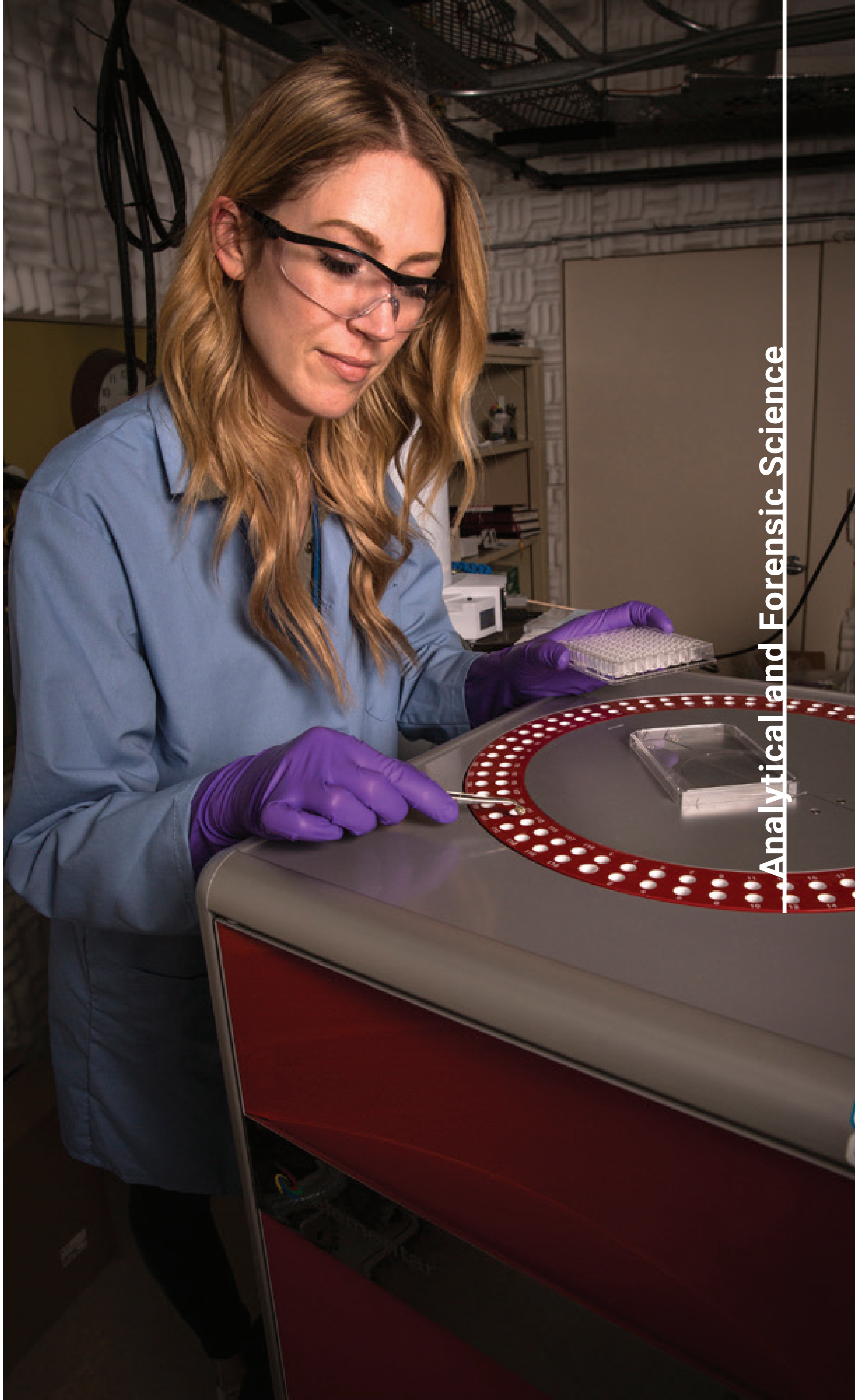
Analytical and Forensic Science

Analytical chemistry, defined by the American Chemical Society as the art and science of determining what matter is and how much of it exists, is one of NACS's foundational competencies. In this area, our goal is to advance forensic science related to CBRNE—chemical, biological, radiological, nuclear, and explosive—threats by developing innovative extraction materials, analytical techniques, and assay methodologies. We approach this technical challenge from the unique perspective of scientists who conduct pioneering research while providing real-world operational support across a broad spectrum of forensic activities, such as analysis of interdicted nuclear materials and proficiency testing for chemical weapons detection. Programmatic and fundamental activities feed one another, ultimately leading to one-of-a-kind instrumentation and exquisitely precise measurements.

In addition to responding to national threats, progress in analytical chemistry offers answers to key questions about the composition, structure, and transport of the matter that surrounds us on Earth, in the Solar System, and beyond. Along with proven applications to pre- and post-detonation nuclear forensics, our world-leading capabilities in inorganic mass spectrometry (MS) enable ground-breaking studies in cosmochemistry, environmental radiochemistry, water resources, and environmental microbial communities.

New Paradigms in Forensic Science

The Forensic Science Center (FSC) is rooted in analyses related to chemical weapons and serves as one of two U.S. laboratories designated by the international Organisation for the Prohibition of Chemical Weapons (OPCW). Research in the FSC has focused on the development of advanced detection capabilities for chemical threats and the exploration of related mitigation approaches. Recently, emphasis has evolved toward the detection of the biochemical signals of human exposure and the development of corresponding countermeasures. Our vision is to expand this approach to the entire CBRNE-phase space and to address critical challenges related to



selective sample collection and detection strategies that can operate remotely or autonomously in the field.

When faced with emerging threats, forensic analysis may no longer be able to depend on libraries of known chemical analytes. In these cases, identification of forensic signatures in complex sample sets will rely on analytes that are pertinent to an investigation but are not yet known. Approaches could include key mass-spectral fragment prediction/recognition, integration of nuclear magnetic resonance (NMR) and isotope ratio MS (IRMS) methods with chromatography/MS, and development of analytical methods and algorithms to evaluate the relationships of various markers within a sample.

The need to understand human exposure responses and signatures for various existing and emerging threats—including chemical weapons, narcotics, pharmaceuticals, and biological or radiological materials—is increasingly pressing. This area of active research in biomarkers pushes the boundaries of biochemistry to develop the robust pharmacokinetic and metabolic knowledge needed to determine exposure levels, timelines, and metabolite signatures.

Forensic science has been under significant scrutiny in recent years, particularly regarding evidence that relies heavily on subjective analysis methods. Recent work at the FSC has focused on protein-based human identification using hair evidence and is now expanding to bone and teeth. This approach as a non-DNA-based identification strategy is creating a new paradigm in traditional forensic science.

Forensics of Terrestrial and Planetary Processes

Fundamental cosmochemistry research focuses on constraining the mechanisms by which the Solar System formed and evolved using mineralogical, geochemical, and isotopic measurements on meteorites and samples returned from international space missions. These activities involve age determinations of primitive meteorite, differentiated asteroid, Martian, and lunar samples, as well as stable isotope measurements on these samples to establish the origin of their constituents. Methods include secondary ion MS (SIMS), bulk sample isotope MS, laser ablation inductively coupled plasma MS, noble gas MS, and resonance ionization MS. NACS places an emphasis on the analysis of short-lived isotopic systems in meteorites and samples returned from the Moon and other extraterrestrial bodies to test theories of the formation and evolution of the Solar System, constrain the time of planetary accretion and differentiation, and evaluate when planetary bodies formed cores, mantles, and crusts. Although these objectives are long-standing goals of the cosmochemical science community, recent developments in instrumentation and techniques will make these measurements possible with enough fidelity to confidently answer these questions.

In order to reliably predict the cycling and mobility of actinides in the subsurface, environmental actinide chemistry research is focused on identifying the dominant biogeochemical processes and underlying mechanisms that control actinide transport in the environment, which is intimately tied to actinide redox behavior. Thus, our research identifies the mechanisms driving surface-mediated reduction of actinides on mineral colloids, the ligands and functional groups responsible for biologically mediated redox transformations, and the factors controlling the stability of intrinsic actinide colloids under environmentally relevant conditions. Cutting-edge research in actinide environmental chemistry requires continued development of novel capabilities that test our conceptual understanding of environmental actinide chemistry. Methods include NMR, electron paramagnetic resonance, NanoSIMS analysis of actinides, and accelerator MS analysis of actinides at sub-femtomolar concentrations.

Water is a critical resource essential for energy and national security. Climate change challenges societies to adapt water resource infrastructure and management (designed and implemented in the last century under different climate) to current and future conditions. For California, the current and predicted climate is and will be warmer with more highly variable precipitation, which will result in a smaller snowpack, earlier melt period, and more severe droughts and flooding. Accurately simulating the impact of this new climate on runoff and water availability requires understanding the secondary adaptation of ecosystems and concomitant changes in water loss to the atmosphere by evapotranspiration. NACS researchers use cosmogenic isotopes to investigate plant water strategies by tracking water through soil and vegetation into subsurface storage.

Smaller snowpack and earlier snow melt effectively reduce snowpack storage from the natural component of the water supply system. Developing new storage solutions in the form of managed aquifer recharge requires a thorough understanding of the future runoff regimes of natural watersheds and the recharge processes enabling deep percolation. For this challenge, NACS has unique expertise in quantifying the process of river recharge under pre-development and managed conditions using stable isotopes, noble gas signatures, and groundwater age dating.

Our system biology team uses the tools of isotope biogeochemistry and molecular biology to study environmental microbial communities, including biofuels, the terrestrial carbon cycle, and environmental remediation. Much of this research hinges on LLNL's pioneering applications of NanoSIMS isotopic imaging for cell and microbial biology, soil biogeochemistry, and plant–microbial interactions. A primary focus is a multidisciplinary effort to understand biotic interactions and energy flow in microbial communities critical to nutrient cycling and sustainable biofuel production. This work has led to the development of several highly recognized isotope-tracing approaches that link microbial identity and function by combining NanoSIMS with other analytical techniques.

We also work at the intersection of microbiology, geology, and chemistry on mechanisms of soil carbon stabilization and environmental biogeochemistry. Because global carbon cycling is ultimately controlled by microscale biogeochemistry, our cutting-edge capabilities in isotopic imaging and electron microscopy can be used in combination with accelerator MS and synchrotron-based methods to provide unique tools to address carbon cycling. The vision to move microbial ecology and molecular biochemistry beyond the current natural history phase—where organisms are simply counted, named, and sequenced—includes focusing on understanding the interactions between cells in space and time and the processes that control interactions and functions of complex microbial communities. Quantitative spatial, chemical, and isotopic analyses developed at LLNL could ultimately play a significant role in the predictive understanding of microbial activities while allowing for engineered solutions for agricultural and natural resources security, the terrestrial carbon cycle, and sustainable biofuels.

Nuclear Detection Technology and Algorithms

Discovery is driven by detection. Our understanding of the universe began with looking at the stars, and ever since Galileo pointed his telescope upward, we have continued to deepen and broaden our view with more capable detection systems. Whenever we improve detection, we learn something new and feed our imagination to probe deeper. The NACS Division is deeply involved in developing new detection systems to study the Solar System, neutrinos, and newly discovered particles while searching for the missing matter in the universe. These detector systems range from small, lightweight detectors for gamma-ray detection to massive, ton-scale detectors to detect neutrinos.

Advances in detection require advances both in the detectors themselves and in the algorithms used to process, analyze, and interpret the data. To develop advanced detection capabilities, researchers draw on a wide range of science and mathematics. Detectors require a medium for photons or particles to interact in, as well as a mechanism to collect the signatures of those interactions and convert them into signals that can be read out for analysis. This requires research in nuclear interactions, material science, solid-state physics, and electronics engineering. Algorithm development can involve research in signal processing, statistics, computer science, and predictive analytics.

Overall, detector development is a multidisciplinary research effort, and the NACS Division is structured to provide the necessary range in expertise needed to work at the cutting-edge of detector and algorithm development and design of test and evaluation schemes to validate and verify detection precision and accuracy. Advances require studying detector materials, simulation and modeling of detector response, readout design, signal processing, and data analytics—all with a clear view toward furthering scientific discovery and solving critical security problems.

Radiation detection in NACS spans a wide range of instruments with seemingly contradictory properties. Measuring new particles at the LHC, probing for weakly interacting massive particles, and studying neutrinos require large systems with incredibly sophisticated algorithms to sort through all the common particles and interactions and find the rare interaction from the particles of interest. Research in new detector materials, including the lowest cost material (water), and readout and analysis algorithms is making significant advances to the field. We address the varied challenges in the detection of radioactivity levels ranging from the ultra-low levels found in the natural environment to the high-count rates exhibited in large quantities of nuclear materials.

In contrast, for gamma-ray and neutron detection, much of the cutting-edge research involves developing smaller, lighter-weight, portable detection systems that bring operators closer to the source with a net gain in sensitivity. Once again, these systems also require sophisticated algorithms to sort through all the clutter from background variations and common, benign radiation sources to find the rare signatures from radioactive materials of concern. Research has enabled hand-held detectors and imaging systems with excellent energy resolution to operate without the cryogenics or excessive electronics and high-power requirements typical of existing standard systems. Lower-temperature systems are being developed to provide even higher-energy resolution for alpha and gamma spectroscopy, along with fieldable cryogen-free cooling systems. New algorithms to support networks and swarms of detectors are being developed, often simultaneously with new approaches to machine learning that significantly improve the computational performance needed for signal processing. The science of fission chains is being advanced along with fast-neutron measurement systems to reveal critical details about fissile material assemblies. In addition, new algorithms and analysis of stellar magnitude variations are being developed that could radically change our understanding of matter and the evolution of the universe.

In many cases, NACS researchers apply new detector developments to advance both scientific frontiers and to improve nuclear security. Neutrino detectors are being developed for neutrino physics experiments and to monitor plutonium production in nuclear reactors. A robust, mechanically cooled gamma-ray detector developed by NACS researchers is orbiting Mercury to study the planet’s composition. New versions are being developed to rendezvous with an asteroid, and a commercial version will soon be available for nuclear security operations worldwide.

ACRONYMS

$0\nu\beta\beta$	neutrinoless double-beta decay
ADMX	Axion Dark Matter Experiment
CARIBU	Californium Rare Isotope Breeder Upgrade
CBRNE	chemical, biological, radiological, nuclear, and explosive
CERN	European Organization for Nuclear Research
CHICO	Compact Heavy Ion Counter
DOE	Department of Energy
DT	deuterium–tritium
EDM	electric dipole moment
EIC	Electron Ion Collider
FRIB	Facility for Rare-Ion Beams
FSC	Forensic Science Center
GRETA	Gamma-Ray Energy Tracking Array
GRETINA	Gamma-Ray Energy Tracking In-beam Nuclear Array
IRMS	isotope ratio mass spectrometry
JETSCAPE	Jet Energy-loss Tomography with a Statistically and Advanced Program Envelope
LBNF	Long Baseline Neutrino Facility
LHC	Large Hadron Collider
LLNL	Lawrence Livermore National Laboratory
LUX	Large Underground Xenon
LZ	name for the merger of the LUX and ZEPLIN collaborations
NACS	Nuclear and Chemical Sciences
OPCW	Organisation for the Prohibition of Chemical Weapons
QCD	quantum chromodynamics
QGP	quark–gluon plasma
MS	mass spectrometry
MSU	Michigan State University
NanoSIMS	nanometer-scale secondary ion mass spectrometer
nEXO	next Enriched Xenon Observatory
NIF	National Ignition Facility
NMR	nuclear magnetic resonance
NSCL	National Superconducting Cyclotron Laboratory
PHENIX	Pioneering High Energy Nuclear Interaction EXperiment
PROSPECT	Precision Oscillation and Spectrum Experiment
RHIC	Relativistic Heavy Ion Collider
SciDAC	Scientific Discovery through Advanced Computing
SIMS	secondary ion mass spectrometry
sPHENIX	successor of the PHENIX experiment
WATCHMAN	WATer CHerenkov Monitor for AntiNeutrinos
ZEPLIN	ZonEd Proportional scintillation in LIquid Noble gases

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