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# CINT 2026

## Center for Integrated Nanotechnologies Strategic Plan



## CINT Mission

The Center for Integrated Nanotechnologies (CINT) is a Department of Energy, Office of Science Nanoscale Science Research Center. CINT operates as a national user facility devoted to establishing the scientific principles that govern the design, performance, and integration of nanoscale materials and structures. Through our Core Facility in Albuquerque and Gateway Facility in Los Alamos, CINT provides access to scientific expertise and advanced capabilities for researchers to synthesize, fabricate, characterize, understand, and scale nanostructured materials into the microscopic and macroscopic worlds. This comprehensive approach provides the greatest potential for nanostructured materials to inspire technological innovation with enduring beneficial impact in energy, environment, human health, and security.

This work was performed, in part, at the Center for Integrated Nanotechnologies, an Office of Science User Facility operated for the U.S. Department of Energy (DOE) Office of Science. Sandia National Laboratories is a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525. Los Alamos National Laboratory, an affirmative action equal opportunity employer, is managed by Triad National Security, LLC, for the U.S. Department of Energy's NNSA under contract 89233218CNA000001.

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## 1 EXECUTIVE SUMMARY

The Center for Integrated Nanotechnologies (CINT) plays a leadership role in the area of nanoscience through its function as a Department of Energy (DOE), Office of Science Nanoscale Science Research Center national user facility. By creating a collaborative community of diverse users and expert scientific staff, CINT fosters high-impact nanoscience discoveries, leads next-generation technique development, and advances the frontiers of knowledge beyond that which is achievable by individual researchers or any single institution.

Each year we revisit our strategic plan to ensure it is focused on meeting the needs of the user community today and into the future. This plan is based upon the extraordinary scientific opportunities involving nanomaterials and quantum phenomena, organized into five challenges representing nanoscience opportunities inspired by CINT users and staff. These challenges, described in Section 3, include: electronic and photonic systems for quantum information science; hybrid material interactions for generation and manipulation of light; assembly of soft and hybrid materials; nanoscale materials science in extreme environments; and machine learning and artificial intelligence in nanoscience. Solving these challenges will have significant impact in important areas such as energy, environment, health, and security.

CINT's role is to enable world-leading science towards realizing these benefits, and our strategic objectives describe what is needed to deliver on this promise. As a vibrant partnership between Los Alamos National Laboratory and Sandia National Laboratories, CINT leverages the unmatched scientific and engineering expertise, as well as special capabilities, of our host DOE laboratories in an Office of Science open-access user facility, benefitting hundreds of researchers annually. We have world-leading scientific expertise in four thrust areas, as described in Section 2, and specialized capabilities to synthesize, fabricate, characterize, and understand nanomaterials in increasingly complex, integrated environments, as described in Section 4.

Building upon these current strengths, we continue to identify capabilities and expertise that the nanoscience community will need in the future and that CINT is well positioned to develop and offer as a user facility. These include an expanding portfolio of our signature Discovery Platforms to include the new quantum NMR platform to study nuclear spin at the smallest length scales; novel synthetic approaches for exquisitely heterostructured nanowires, nanoparticles and quasi-two-dimensional materials; metamaterials for photonics and optoelectronics; nanomechanics for fundamental understanding of the effects of defects/crystal distortions on the mechanical properties; ultra-fast and ultra-high resolution spectroscopic techniques of nanomaterial dynamics; in-situ microscopies that provide real-time, spatially-resolved structure/property information, advanced simulation techniques, and multi-scale theory for interfaces and dynamics. CINT has significant expertise in quantum information science, offering capabilities in measuring single spin electron spin resonance and nuclear magnetic resonance in quantum dots and defect centers in quantum materials, and electronic and photonic capabilities for creation of novel qubits.

CINT's transformational and supporting foundational capabilities are made available to the widest possible number of qualified users via a comprehensive communications and outreach effort combined with an efficient user proposal, peer-review, and project management system.

We target research leaders in diverse institutions, early career scientists, and innovators in nanotechnology industries to create an expanded national nanoscience user community of the future.

## 2 INTRODUCTION

The Center for Integrated Nanotechnologies (CINT) is a Department of Energy (DOE), Office of Science Nanoscale Science Research Center (NSRC) operating as national user facility. As a vibrant partnership between Los Alamos National Laboratory (LANL) and Sandia National Laboratories (SNL), CINT leverages the unmatched scientific and engineering expertise, as well as special capabilities, of our host DOE laboratories, creating a unique user facility environment among the NSRCs. Our users and staff conduct research projects within and across the *Core Facility* in Albuquerque, NM, and the *Gateway Facility* in Los Alamos, NM (Figure 2.1). By creating a collaborative community of diverse users matched to expert facility scientists with advanced capabilities, CINT fosters high-impact nanoscience discoveries, leads next-generation technique development, and advances the frontiers of knowledge beyond what is achievable by individual researchers or any single institution.



Figure 2.1 CINT Core Facility (left) and Gateway Facility (right).

Our overarching goal is to be the national resource for research expertise and advanced capabilities to synthesize, fabricate, characterize, understand, and integrate nanostructured materials. By developing innovative systems with unprecedented functionality we aim to inspire revolutionary nano-enabled technologies.

In order to achieve this goal, CINT has the following six strategic objectives:

1. CINT will be a nationally recognized leader in nanomaterials.
2. CINT will develop unique experimental and theoretical capabilities to synthesize, fabricate, characterize, and understand nanoscale materials in increasingly complex environments.
3. CINT will continue to develop new generations of Discovery Platforms inspired by materials challenges to provide a window into nanoscale and quantum phenomena.
4. CINT will invest to provide foundational capabilities and commercial instrumentation necessary for internationally competitive nanoscience research.
5. CINT will operate safely and effectively by aligning capabilities and capacity with the ever-evolving user demand.
6. CINT will increase the diversity and breadth of our national user community, foster high-impact science, and encourage intellectual risk in basic and use-inspired research.

Together, these CINT strategic objectives support and align with the U.S. DOE’s strategy to deliver the scientific discoveries and major scientific tools that transform our understanding of nature and strengthen the connection between advances in fundamental science and technology innovation. CINT supports these objectives by: (1) Conducting discovery-focused research, (2) Providing the nation’s researchers with world-class scientific user facilities, and (3) Leveraging the national laboratory system as well as partnerships with universities and industry to conduct mission-focused research.

We have world-leading scientific expertise and specialized capabilities to synthesize, fabricate, characterize, and understand nanomaterials in increasingly complex integrated environments. Our expertise is organized in four scientific thrust areas:

- **Quantum Materials Systems (QMS):** Understanding and designing nanomaterials to create new functionalities based on quantum effects that span multiple length scales (from nm to mm).
- **Nanophotonics and Optical Nanomaterials (NPON):** Discovery, synthesis, and integration of optical nanomaterials; exploitation and characterization of emergent or collective electromagnetic and quantum optical phenomena, from nanophotonics and metamaterials to quantum coherence.
- **In-situ Characterization and Nanomechanics (ICNM):** Developing and implementing world-leading capabilities to study the dynamic response of materials and nanosystems to mechanical, electrical, radiation, or other stimuli.
- **Soft, Biological, and Composite Nanomaterials (SBCN):** Solution-based materials synthesis and assembly of soft, composite, and artificial bio-mimetic nanosystems.

In addition, we leverage the strengths of our host DOE laboratories through the alignment of the CINT 2026 strategic plan with the LANL and SNL missions, capabilities, and strategic investments (Figure 2.2). For example, through the exploration of materials and integration, we support the LANL “Materials for the Future of Los Alamos” scientific pillar which seeks to establish the design principles, synthesis pathways, and manufacturing processes for advanced and new materials to intentionally control functionality and performance prediction relevant to national security missions. Similarly, the SNL Strategic Plan includes explicit objectives in support of the nanoscience capability, Materials Science Research Foundation, Advanced Science and Technology Program, and multiple crosscutting research challenges involving the creation, understanding, and integration of advanced nanomaterials in technologies for national security applications.

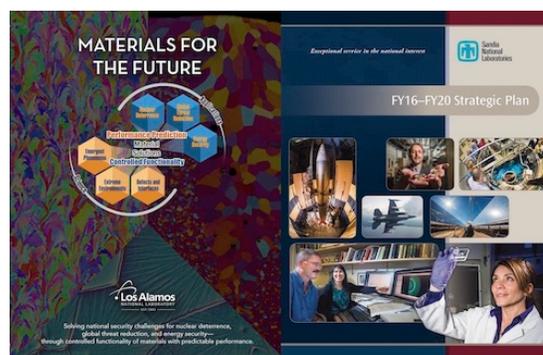


Figure 2.2 LANL (left) and SNL (right) strategic plans.

The coordination of SNL, LANL, and CINT strategic plans leads to very visible benefits for all three entities. Researchers from LANL and SNL continuously request access to CINT’s capabilities and expertise through User project collaborations. Over the past three years CINT has hosted over 900 host facility researchers, reinforcing the message that the laboratories value our Center, and

that CINT closely partners with its host institutions to deliver high impact contributions to the labs' core missions. These host laboratory projects are funded by a wide variety of sponsors including, but not limited to, the Department of Energy (including Laboratory Directed Research and Development (LDRD)), the National Nuclear Security Administration, the Department of Defense, the National Science Foundation, Homeland Security, and the National Institutes of Health.

The exemplary science and technology that come from the CINT staff and collaborations with host lab and external users has received recognition by renowned sources. Most notably, CINT has won four R&D 100 awards in recent years. The R&D 100 Awards identify and celebrate the top technology products of the year spanning industry, academia, and government-sponsored research. Our R&D 100 winners include:

- Binary Solvent Diffusion for Fabrication of Large Nanoparticles Supercrystals (H. Fan, D. Huber, W. Luk, E. Vreeland, et al., 2020)
- LAMMPS Molecular Dynamics Simulator (M. Stevens, et al., 2018)
- Transceiver for Quantum Keys and Encryption (R.M. Camacho, J. Urayama\*, P. Davids\*, et al., 2016)
- Stress-induced Fabrication of Functionally Designed Nanomaterials (H. Fan\*, T.S. Luk, I. Brener, et al., 2016)<sup>1</sup>

These achievements illustrate the ever-growing potential for CINT to amplify the exceptional nanoscience strengths of our host DOE laboratories. Over 20 CINT scientists have also been named fellows of professional societies, including societies such as the American Physical Society and the American Chemical Society.

Finally, we note the special role the DOE Office of Science user facilities have in supporting key objectives in 2016 National Nanotechnology Initiative Strategic Plan. Objective 3.3 is to provide, facilitate the sharing of, and sustain the physical and cyber R&D infrastructure, notably user facilities and cooperative research centers. This requires: (3.3.1) Establishing regular mechanisms to determine the current and future infrastructure needs of users and stakeholders of these facilities and centers, and (3.3.2) Developing, operating, and sustaining state-of-the-art tools, infrastructure, and user facilities, including ongoing investment, staffing, and upgrades.

Nanomaterials integration is key to numerous extraordinary scientific and technological challenges. Solving such challenges would unlock great possibilities for innovative technologies and have widely recognized impact in nationally important areas such as energy, environment, health, and security. We describe five representative challenges in Section 3, followed by the

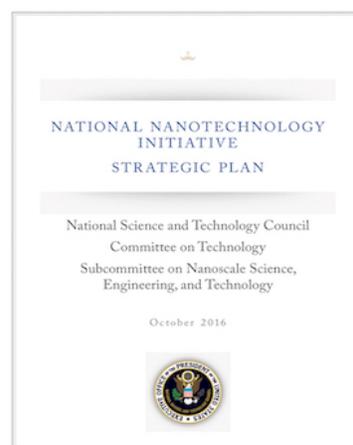


Figure 2.3 NNI Strategic Plan.

<sup>1</sup> CINT users are marked with an asterisk.

requisite expertise and capability development in Section 4, respectively. We also see new opportunities to refine the role of a user facility to meet the needs of a highly networked, international community.

Our vision for CINT in the year 2026 and dedication to continued excellence as a DOE Office of Science user facility ensure that CINT will transform our understanding of nature and strengthen the connection between advances in fundamental science and technology innovation.

### 3 SCIENCE VISION: CHALLENGES IN NANOMATERIALS INTEGRATION

Nanoscale materials exhibit extraordinary physical, chemical, and/or biological properties. Isolated or individual nanoscale materials are scientifically interesting, but they rarely make significant technological impact. Building blocks comprised of individual nanoscale materials are commonly integrated with other materials into architectures that enhance their properties (up-scaling) or lead to new emergent behaviors. By surveying the integrated environments of greatest potential impact, and by developing a fundamental understanding of the principles that govern the integrated properties and behaviors, we can capitalize on the greatest potential for nanomaterials to have an enduring impact on scientific and technological innovations. Nanoscale integration has the potential to revolutionize the way we live, in the same way that the development of the semiconductor-based integrated circuit (IC) did. The development of the IC required the capability to integrate a large number of resistors, capacitors, diodes, and transistors on a single platform. Once the IC was developed, it enabled countless innovations. CINT envisions similarly transformational technologies will ultimately emerge from nanomaterials integration.

Understanding the principles of nanomaterials integration has been the central theme of CINT since its inception. The CINT2026 Strategic Plan builds upon this foundation by illustrating five representative, forward-looking integration challenges inspired by the nanoscience community. These integration challenges enable us to identify the capabilities that the nanoscience research community will need to realize the smart integration of nanomaterials into innovative and competitive technologies. To accomplish these challenges, CINT will bring together expertise and capabilities across our four Science Thrusts. An underlying theme of all five challenges is a fully integrated feedback loop of synthesis, fabrication, characterization, and modeling, shown in Figure 3.1 (and also described in more detail on the NSRC Portal<sup>2</sup>), that will allow the nanoscience community to realize and develop the ability to predict and a priori design unprecedented materials functionalities and innovative systems.

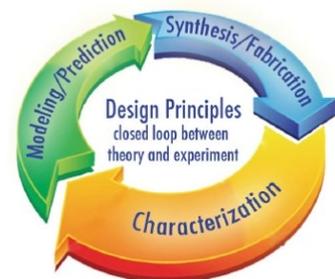


Figure 3.1 CINT leverages its diverse scientific community in an on-going cycle of scientific discovery.

#### 3.1 Electronic and photonic systems for quantum information sciences

##### Scientific opportunity

The BES Roundtable on “Opportunities for Basic Research for Next-Generation Quantum Systems”<sup>3</sup> outlines a number of priority research opportunities:

- Advance artificial quantum-coherent systems with unprecedented functionality for QIS
- Enhance creation and control of coherence in quantum systems
- Discover novel approaches for quantum-to-quantum transduction
- Implement new quantum methods for advanced sensing and process control

<sup>2</sup> <https://nsrcportal.sandia.gov/>

<sup>3</sup> [https://science.osti.gov/-/media/bes/pdf/reports/2018/Quantum\\_systems.pdf](https://science.osti.gov/-/media/bes/pdf/reports/2018/Quantum_systems.pdf)

Advances in each of these Quantum Information Science (QIS) research opportunities requires nanoscale integration.

At CINT we have expertise in materials growth, synthesis, characterization, fabrication, and integration. Over the next five years, we will address integration challenges involving deterministic control of quantum defects, sensing using quantum systems, transduction between qubits, and application of QIS measurements to fundamental nanoscience problems. CINT is well positioned to lead these scientific directions and support users in the QIS community.

### Positional science and capabilities

CINT has performed numerous trail-blazing works in electronic and photonic systems. CINT QIS capabilities are drawn from the Quantum Materials Systems Thrust and Nanophotonics Thrust and are a subset of those thrust activities. Materials are critical for QIS, and CINT has unique capabilities in high mobility GaAs MBE, SiGe nanowires, hybrid semiconductor/2D materials, and complex oxides. The QIS effort also benefits from expertise in synthesis of non-blinking giant quantum dots and chemical functionalization of nanomaterials. Ion implantation, including both broad beam and focused ion beam implantation for high spatial resolution, in materials such as diamond and silicon carbide has been used to create color centers that can be used as qubits, quantum sensors, and transduction elements. Quantum defects can also be synthesized in a bottom-up process through chemical functionalization of nanomaterials, and using carbon nanotubes with defect sites in a plasmonic structure we demonstrated indistinguishable photons at telecommunications wavelengths. At CINT we have expertise in controlling single spins with electron spin resonance (ESR) and nuclear magnetic resonance (NMR), measuring optical properties of defect centers in quantum materials, and manipulating qubits using electronic and photonic techniques. We have a wide range of characterization tools for QIS and quantum materials including electrical transport in dilution refrigerators, optical spectroscopy for single photon, and photoluminescence studies and scanning tunneling microscopy.

CINT materials growth and synthesis capabilities are a key enabler for future QIS technology. We have demonstrated high performance semiconductor systems such as high mobility GaAs heterostructures, SiGe nanowires for energy applications, large bandgap GaN/AlGaN materials and hexagonal BN. We have capabilities for integration and characterization of disparate 2D materials and 2D materials with traditional semiconductors. Perovskite materials have unique and tunable electronic and optical properties and represent long-term economical approach to make materials with quantum properties more widely available. We also have high quality crystal growth of complex materials that can be used to study topological and strongly interacting materials. This wide range of materials capabilities enables many of the future challenges in single defect studies, quantum sensing, quantum transduction and other QIS systems.

CINT has a collection of femtosecond laser systems with complementary capabilities. In our laboratories, several amplified femtosecond Ti:sapphire systems generate pulses from terahertz (THz) to soft x-ray frequencies. We use these capabilities to investigate nearly every class of quantum material, including 2D-TMDCs, transition metal oxides (TMOs), graphene, and Dirac materials. CINT also developed and applied advanced theoretical tools, including real-time

density functional theory and dynamical mean-field theoretical methods, to tackle correlated quantum materials.

Nanoscience integration will enable quantum computing techniques to be used for new measurement capabilities of nanoscale systems. Furthermore, we can draw upon capabilities in our host laboratories for single ion implantation of color centers in diamond, and quantum information theory.

## Toward the future

### Deterministic Placement and Integration of Defects

Can we create individual quantum systems with tailored quantum properties for qubits, sensing and single photon structures?

We will establish new CINT-based infrastructure capable of addressing the needs for: (1) Deterministic creation of atomic defects capable of mimicking trapped ions in a wide variety of host materials; (2) Understanding the correlation between the atomic structure of defects and their quantum functionalities; and (3) Integration of defects into photonic/electronic devices.

To achieve atomic defects that mimic trapped ions, we will employ two complementary strategies: top-down ion implantation technologies and a bottom-up approach for chemical quantum-defect implantation. For the top-down strategy, a new focused ion beam capability will be established at the SNL Ion Beam Laboratory to achieve ion implantation with a variety of atomic species and <20 nm spatial precision. For bottom-up chemical implantation, covalent functionalization chemistry will be combined with lithography tools such as dip-pen nanolithography (DPN). DPN has been uniquely advanced for the deterministic placement of nanocrystals and will be expanded here for deterministic chemical functionalization with nanomesoscale spatial resolution.

Novel in-situ/operando single-defect optical characterization capabilities will be integrated with these ion and chemical defect implantation capabilities to achieve an unprecedented, one functional defect per one site, 100% deterministic defect creation capability. The fundamental understanding attained in correlating atomic structure with defect capabilities will further enhance the capabilities of deterministic creation of quantum defects by revealing a path toward tailoring quantum behaviors of the defects. Finally, we will incorporate quantum defects into the prefabricated electronic/photonic devices with nanometer scale precision by using both top-down and bottom-up strategies. Through these experiments we will establish our deterministic defect creation strategies as the perfect toolset for eliminating the “integration bottleneck” impeding defect-driven QIS research.

### Quantum sensing

Can we integrate quantum systems with nanomaterials and devices to achieve unique new characterization tools for nanoscale systems?

Nitrogen-vacancy centers in diamond combine extreme magnetic field sensitivity with nanoscale resolution. With optical initialization and readout and microwave manipulation of energy levels, NVs are being used to implement the Quantum-Sensed NMR Discovery Platform. By placing nanoscale systems on diamond substrates, NMR will be demonstrated on colloidal nanoparticles, 2D materials, and donors in semiconductors. Working with CINT users, we will integrate user nanomaterials with the NMR sensing capability. Using focused ion beam techniques, color centers can be placed with nanometer precision. Such small implant regions enable enhanced resolution techniques at length scales much smaller than the diffraction limit of light. With nanoscale implantation, color centers can be incorporated into optical resonators, metamaterials, and more sophisticated quantum devices.

Color centers have unprecedented capabilities for sensing phenomena at the nanoscale, but to fully realize the promise of quantum sensing, new approaches are needed for coupling nanomaterials with quantum sensors. Using top-down lithography approaches we are bringing nanomaterials to the diamond by placing nanomaterials on diamond substrates patterned with color centers. In the future, CINT will investigate alternate approaches that combine our unique ion implant capabilities with diamond membranes and scan probe systems providing the ability to bring the diamond to the nanomaterial.

While nitrogen-vacancy centers in diamond represent a core quantum sensing technology, investigating other quantum defects in diamond, other wide bandgap materials, and quantum materials will enable the next generation in quantum sensors. This will include Deterministic Placement and Integration of Quantum Defects (see previous section) and

- Sensing regions with several different types of color centers
- Combined color center sensors with nanoelectronics materials such as GaN and SiC
- New color centers with band structures optimized for improved sensitivity to local electric fields, temperature, and strain.

## Transduction

Can we develop tools for coupling two different quantum systems?

Qubit technology has been developed in a number of physical systems such as superconductors, semiconductors, atomic systems, quantum defects, and topological materials. To make a good qubit, many scientific and technical issues must be solved. Qubit performance always involves trade-offs. One example is long coherence times can be achieved when a quantum system is isolated, but creating multiple interacting qubits can be very difficult in an isolated system. Qubit transduction is an approach where nanoscale elements enable communication of quantum information between different qubit systems. When transduction techniques are developed, QIS researchers will be able to incorporate strengths of different qubit systems while minimizing their weaknesses. Transduction techniques will also be critical as new materials and new qubits get incorporated into more mature qubit technologies. At CINT, integration of different nanoscale

systems can provide the tunable elements needed to transform quantum information. Some examples include opto-mechanical system, tunable superconducting circuits, valley pseudo-spin in 2D semiconductors, and deterministic quantum defects.

#### Materials for QIS

Can we develop material architectures for generation and manipulation of quantum information?

Quantum states can be generated and manipulated in various nanomaterials including superconductors, atomically thin semiconductors, semiconductor heterostructures, and complex materials. However, existing material systems have not been utilized for an integrated quantum information system to generate, compute, and transduce in a system. Absence of silver bullets for quantum information science implies that heterogeneous integration of various quantum materials at nanoscales is imperative for quantum information science and implementation of the quantum technologies. Diverse portfolio of materials for quantum information science, available at CINT, contributes to wide ranges of quantum science disciplines, such as spin-based, photon-based, charge-based, and topological carrier-based quantum systems. At CINT, integrated suites of theoretical prediction/explanation and experimental demonstration of quantum materials at nanoscales have been developed. Demonstrations of low-dimensional high-mobility charge carrier channel in semiconductor quantum wells, heterostructures composed of 2D materials and semiconductor nanowires, strongly correlated heavy fermion materials, quantum dots/nanowires/2D materials embedded in device architectures open up novel opportunities to make synergies between nanoscience and quantum information science.

### 3.2 Hybrid material interactions for generation and manipulation of light

#### Scientific opportunity

Structured hybrid materials can be engineered to have novel photonic properties that emerge only as a result of multi-material interactions and can also include pre-designed properties for new photon generation and manipulation pathways. CINT is advancing the understanding and application of these revolutionary hybrid systems by addressing the most significant open questions surrounding the control, integration, and enhancement of the photonic response of two classes of materials and their associated assemblies: (1) materials and structures that control and modify electromagnetic energy (plasmonics, metamaterials), and (2) materials and assemblies that actively generate and harvest electromagnetic energy.

#### Positional science and capabilities

CINT's foundations to lead in this area rest on our multidisciplinary capabilities for generation of unique photonic materials and their characterization with powerful spectroscopic tools, paired with a growing ability to define material interactions across multiple length scales and degrees of complexity. Specifically, we are international leaders in developing exceptional optical nanomaterials with highly tunable and controlled photon emission properties. Examples include proprietary non-blinking/non-photobleaching quantum dots that now span the green visible to the infrared and our doped single-wall carbon nanotubes whose defect-state emission spans

1150 to 1570 nm with single photon purity. In both cases, the novel quantum emitters are chemically engineered (through heterostructuring and surface chemistry, respectively) to provide single and multi-photon control. At the same time, we have been a world leader in the area of metasurfaces/metamaterials (THz to near-infrared) for nearly a decade. We lead the world in all-dielectric metamaterials and have made key contributions to metallic/plasmonic and dynamic metamaterials. We envision advances in the intentional combination of the optical nanomaterial and metamaterial “building blocks.” To this end, pioneering efforts in nanomaterials integration and assembly include innovative application of scanning probe techniques, such as dip-pen nanolithography, to directly place quantum emitters into metamaterial and photonic/plasmonic cavity structures with nanoscale precision, as well as a custom-built AFM nanolithography system that will use nanopipette delivery of liquid “ink” and possess a coupled optical microscope. The latter will enable *in situ* real-time characterization of delivered nanocrystalline quantum emitters, or even of emissive defect states created upon delivery of chemical reactants that are known to produce optically active defects in 1D materials to, e.g., 2D materials. We are also developing approaches for creating hybrid and hierarchical functional systems using colloidal and molecular self-assembly, including advanced framework chemistry, toward emergent and multifunctional optical, optoelectronic and quantum optical properties.

CINT's position in these two fields—optical nanomaterials and photonic metamaterials—is enabled by advanced materials synthesis (supported by custom automation and microfluidics techniques) and access to world-class nanofabrication and epitaxial growth facilities. Knowledge-driven discovery of new materials is enabled by world-class spectroscopic characterization and theory. The former includes ultrafast tools providing femtosecond temporal resolution across THz to soft X-ray energies. Paired with the broadest continuous excitation range available for Raman spectroscopy (near-IR to UV) and state-of-the-art tools for microscopic imaging, spectroscopy, and dynamics measurements of single nanoelements, CINT's abilities for optical characterization of nanomaterials are unmatched, especially since many of these measurements can be done at low temperatures and under high magnetic fields. Our first-principles density functional theory (DFT) simulation capability encompasses nearly all flavors of electronic structure codes for understanding electronic, optical, and vibrational properties of complex materials. In particular, the LANL-owned nonadiabatic excited-state molecular dynamics capability excels at modeling the largest systems accessible for nonlinear and time-dependent spectroscopy. Theory efforts are further founded in DFT and classical electromagnetic theory simulations for metamaterials modeling and design.

### Toward the future

We seek to move beyond nanoscale or quantum size effects to capitalize on a new ability to manipulate light and enhance light-matter interactions afforded by precision integration of different material classes into hybrid material systems. Drawing on our strengths, CINT will address the following opportunities in hybrid photonic materials as we move forward.

#### *Generation and active manipulation of novel emitting states and photon correlation statistics*

Accessing new emission regimes of expanded wavelengths, enhanced quantum yields, and tunable or selectable photon statistics and dynamics requires defining interactions in terms of

the relative placement and orientation of materials within a hybrid structure, while also controlling the hybrid composition over multiple dimensionalities. CINT will expand the hybrid materials community's ability to generate and actively manipulate novel emitting states and photon correlation statistics by addressing the following challenges:

- Identifying and realizing candidate materials that are likely to generate targeted optical behaviors from hybrid interactions, such as tailoring of plasmonic interactions aimed at enhancing biexciton emission or overcoming decoherence (Figure 3.2).
- Creation of multi-component systems with the appropriate interaction geometries for a desired functionality, such as harnessing metamaterial interactions with dopant states of emitters for directional emission, enhanced coupling to photonic waveguides, or spin state control/readout.
- Generation of interactions across multiple length-scales that lead to emergent and collective optical responses. Examples include use of 'soft' responsive systems to modulate coupling between embedded optical emitters for on-demand behaviors.

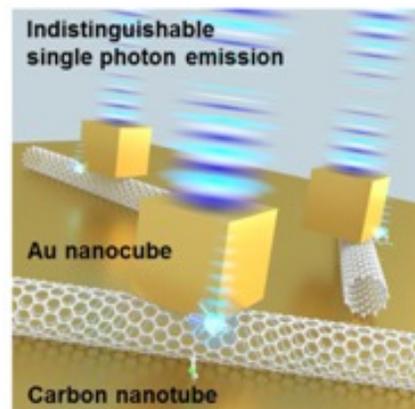


Figure 3.2 By coupling quantum emitters to plasmonic cavities, new or enhanced "hybrid" functionality can be achieved, e.g., SWCNT defect-state emission can overcome decoherence. The "by-chance" interaction will be made deterministic by addressing challenges in rational integration.

To develop new functional materials and to advance our existing suite of quantum/optical nanomaterials, predictive and high-throughput theoretical techniques will be established (see below). To realize the identified candidate materials, new synthetic strategies for composition, phase, size and shape control at the atomic and nanoscale, including the direct synthesis of hybrid nanostructures, will be developed. This will be aided by advances in synthesis automation, *in situ* diagnostics, and, ultimately, autonomous feedback control guided by machine learning algorithms. Creation of multi-component systems that comprise disparate material types, e.g., a chemically prepared 0D quantum dot, 1D nanotube, or 2D transition metal dichalcogenide (TMD), and a 'hard-fabricated' optical cavity or nanoantenna, will need new ways to both deterministically and rapidly integrate one with the other. Finally, to realize functional and dynamic interactions across multiple length scales, programmable and stimuli-responsive polymers or other multi-scale/multi-component templates will be developed to harness the responsive and highly tunable nature of bio-inspired systems.

#### *Active, multifunctional plasmonic and metamaterial interactions*

Hybrid materials interactions have significant potential for establishing new functionality and enhanced manipulation of the medium in which light is generated, harvested, or propagated. Hybrids enable a move from passive to active plasmonic and metamaterials, and form a basis for new concepts including metamolecules (in which the collective interactions of individual metamaterial elements or atoms create new function), "plasmonics on demand" (where localized materials interactions automatically generate desired resonances in optimized locations), and tailored and enhanced optical nonlinearities in metamaterials coupled to different

materials (including quantum systems). Unprecedented multifunctionality will result, giving simultaneous control of polarization states, beam steering, and focusing; integrating perfect absorption of light directly into optoelectronic architectures; or ultimately integrating hybrid metamaterials directly with emerging concepts in emitting materials. For example, we will use hybrid materials (e.g., doped CNTs integrated with FET or plasmonic devices) as single photon emitters, and then build upon this to control many photons or spins via soft/hard (TMD/metasurface) and soft/soft (quantum dots and metal-organic frameworks) material interactions. To realize this extraordinary multifunctionality, CINT will utilize our integrated efforts in synthesis, characterization, and modeling to address the following:

- Devising non-traditional plasmonic systems (e.g., graphene hybrids or emerging epitaxial oxides) that cannot be accessed with more traditional noble metal approaches.
- Designing multifunctional metamaterial behaviors through metamolecule concepts (shown in Figure 3.3).
- Generating hybrid interactions coupled to metamaterial architectures to provide active/dynamic control and tuning of enhanced metamaterial response and their optical nonlinearities.

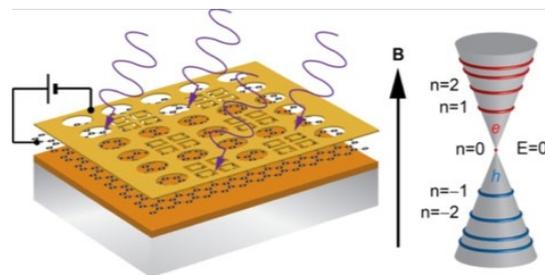


Figure 3.3 Integrated meta-molecule-based metamaterial.

*Meeting the modeling challenge:* CINT's ultimate goal in this area is to create hybrid materials by design. This will entail significant advances in predictive modeling, including machine-learning enabled. In particular, the state-of-the-art must be dramatically advanced in such areas as electronic structure, dynamics, environment, and interfacial interactions at length-scales between the molecular and macroscopic. To meet the modeling challenge, CINT will pursue the following opportunities:

- Develop a predictive capability for designing new optical functionality arising from materials interactions, such as between plasmons in metallic systems (e.g. metal nanoparticles, topological semimetals) and excitons in semiconductor nanoemitters.
- Understand materials coupling mechanisms and identify the most interesting and promising materials interactions, both in terms of composition and interaction geometries, to pursue as routes to novel optical behaviors.
- Develop new theoretical concepts capable of optimizing electronically active networked structures by accessing the middle-length scales of significance for understanding integrated hybrid behaviors and obtaining targeted optical responses.

CINT's current expertise and capabilities in hybrid materials provide a strong foundation for pursuing the above questions. To address the full range of effort in this area CINT will expand its materials generation capability to include new techniques capable of reliable placement of optical nanomaterials with nanometer precision or deterministic creation of new emissive states. Additionally, our strengths in single-nanoparticle spectroscopic characterization have been significantly enhanced by adding capabilities for single nano-element Raman and magneto-optical spectroscopy while expanding our capabilities to include single-photon counting and

ultrafast microscopy techniques at wavelengths longer than the near-IR. CINT will also bring the full strength of our integrated efforts in synthesis, characterization, and modeling to bear on these issues. Model development will work hand-in-hand with experiment in this rapidly expanding field, to allow us to establish the most relevant test systems for validation of predictive models. There is significant new opportunity for developing robust approaches to model exciton-plasmon coupling, integrate quantum materials with metamaterials, and pioneer the area of “phononics” (manipulation of phonons and phonon coupling phenomena).

### 3.3 Assembly of Soft and Hybrid Materials

#### Scientific opportunity

Nanoscience has significantly advanced over the past two decades, but many issues remain with respect to scaling-up nanoscale functions to impactful applications. One approach to this challenge involves the hierarchical assembly of soft nanomaterials (e.g., polymers, gels, and biomolecular materials) across length scales while imparting and regulating the interfacial interactions, dynamics, and metastabilities. CINT is advancing the forefront of soft matter assembly through the synthesis, characterization, and theory of soft nanomaterials with a unified goal of integration into functional architectures. In the coming years, CINT will build on this foundation with investments in science and capabilities in:

- Enhanced and automated control of nanomaterial synthesis
- Precise control of multiscale assembly of polymer, hybrid, and composite nanomaterials
- Understanding how interfaces control assembly processes through multimodal characterization and multiscale modeling

#### Foundational science and capabilities

Soft nanomaterials science at CINT is supported by our multidisciplinary science and capabilities in the synthesis of soft and biomolecular building blocks, active and directed self-assembly processes, high-resolution characterization tools, and multiscale theory and simulations. In synthesis, CINT has capability in the custom design and synthesis of structural and functional biomolecular nanomaterials for integration in soft and hybrid composites. CINT also maintains capabilities in the synthesis and functionalization of engineered nanoparticles, such as magnetic nanoparticles, fluorescent metal nanoclusters, and plasmonic nanoparticles. Central to this effort is CINT’s Microfluidic Discovery Platform™ that permits the synthesis, functionalization, and real-time characterization of nanoparticles and composites that feeds directly back to synthesis. We are also pioneering novel approaches to the assembly of nano-building blocks into functional complexes. These include pressure-driven assembly, motor-driven assembly, and stimuli-responsive assembly and self-organization.

CINT maintains world-leading capabilities for the characterization of the spatial distribution and temporal dynamics of soft and biological nanomaterials within complex environments. This includes the ability to introduce fluorescent reporters for super-resolution optical microscopy and multi-dimensional tracking. The single molecule/particle tracking microscope at CINT enables visualization of complex dynamic systems, for example, tracking the three-dimensional

movement of receptors on a biomembrane in real-time. Further, super resolution optical microscopy permits imaging fluorescently labeled biomolecules/nanoparticles with a resolution of 10-20 nm. CINT has established a cryogenic electron microscopy (cryo-EM) imaging suite that includes a Scios 2 dual-beam SEM/FIB with a dedicated cryo-cooled stage and Talos L120C TEM with a camera optimized for low keV imaging. Specialized transfer equipment permits cryogenic focused ion beam (FIB) milling, lift-out and transfer to the TEM. CINT will also be establishing a new capability in cryogenic correlated light and electron microscopy (cryo-CLEM) that combines the strengths of the two modalities for characterizing structure-function relationships using a single sample. This new capability will also leverage core capabilities in the engineering and synthesis of biomolecular materials with fluorescent nano-tags, as well as the synthesis of metal nanoclusters and colloidal quantum dots as fluorescent reporters in cryo-CLEM.

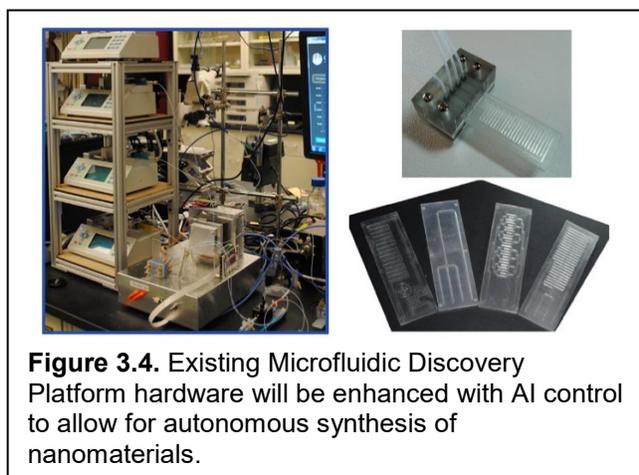
CINT's synthesis and characterization capabilities are underpinned by developing a fundamental understanding of the interactions that occur among nanoscale components, and how these interactions regulate their hierarchical assembly and overall functionality. CINT continues to develop methods to model polymer nanocomposites, biomolecules and biomolecular materials and nanoparticle self-assembly using classical density functional theory, self-consistent field theory, free energy theory, Monte Carlo simulations and molecular dynamics simulations. One of our main tools is the open source, Sandia-developed molecular dynamics package, LAMMPS, which is uniquely capable for simulating the dynamics of nanoparticles from the single particle level to complex systems containing a billion atoms.

#### Toward the future

The translation of nanoscience into useful technologies requires the integration of nanoscale materials/components that translate unique nanoscale physics into emergent functionality at the macroscale. Here, Nature provides powerful inspiration and models such as the lightweight but incredibly strong shell of abalones and adaptive color changing of cephalopods. These examples underpin the unparalleled structural and functional complexity that is achievable through soft nanomaterial integration. Moreover, the immense possibilities that exist in biology provide an underlying confidence and direction for the design and development of considerably more modest, but increasingly complex and functional systems with soft and hybrid (biotic/abiotic) nanomaterials. CINT has established a strong scientific foundation in the integration of soft nanomaterials that will be broadened over the next few years with a focus on: (1) enhanced and automated control of nanomaterial synthesis; (2) precise control of multiscale assembly of polymer, hybrid, and composite nanomaterials; and (3) understanding how interfaces control assembly processes through multimodal characterization and multiscale modeling.

### *Enhanced and automated control of nanomaterial synthesis*

Nanoparticle synthesis remains, in many ways, as much art as science. Because we are unable to effectively predict how new systems may behave, we typically execute repeated reactions while adjusting parameters like temperature and reagent concentrations between attempts. Guided primarily by human intuition, this is a vastly time- and resource-inefficient process. New advances in AI offer us two potentially transformative opportunities. First, we will integrate AI directly into our characterization and synthesis hardware to enhance the Microfluidic Discovery Platform (Figure 3.4) to use real-time characterization data to allow for computer-controlled synthesis, where the AI implements informed, continuous adjustments using reinforcement learning methods to the reaction conditions in order to discover ideal synthetic conditions. Second, AI control can actually uncover the fundamental processes of nanoparticle formation. For example, implementing an Equation Learner system, does not just find mathematical approaches to optimization, but produces the equations that govern reactivity. We can use these equations, coupled with our existing understanding of the reactivity of the species in a reaction, to determine the fundamental mechanistic steps of a nanoparticle reaction.



The goal is to implement AI methods to design stable, consistent reactions that yield a desired product with enhanced reproducibility, which fundamentally changes the synthesis approach. This approach to nanomaterials synthesis and assembly can be applied to new systems that have never been synthesized before, or to tune the synthesis of known materials to produce novel properties. By combining foundational AI technologies with principles of materials chemistry, physics and engineering to design optimal chemical reaction networks that will improve reaction conditions, we can dramatically decrease the time to design reactions, perhaps by as much as 100-fold, reducing a reaction that previously could have taken a year to design to days. We will use AI search techniques to synthesize materials with targeted properties, and we will develop physics-informed neural networks to extract fundamental physical and chemical equations hidden in nanomaterial synthesis data. AI research will be carried out to solve critical bottlenecks in real-time control over synthesis of new nanomaterials. We expect that the AI-centric research will lead to on-the-fly learning of potential connections between the synthesis parameter space and growth mechanisms, predictive control over synthesis with a feedback loop, and high-throughput analysis of real-time materials characterization data.

### *Synthesis, assembly, patterning and theory of soft nanomaterials*

The fundamental building blocks of synthetic, soft nanomaterials are, in general, structurally and chemically simple in contrast to their biological analogs (i.e., proteins). Controlling the size, shape, and crystal structure of nanoparticles is readily achievable in many systems. However, imparting precise chemical composition to soft nanomaterials and defining functionality at the

surfaces of nanomaterial building blocks remains an important science challenge with respect to directing their assembly and organization across multiple length scales and dimensions. This challenge extends to synthetic (e.g., polymers) and biomolecular nanomaterials (e.g., peptide/proteins) in which the building blocks encode the information necessary to instruct assembly and integration into functional architectures.

CINT will focus on developing capabilities for the synthesis of sequence-controlled nanostructured polymers, colloidal and polymer-grafted nanoparticles, and block copolymers. Complementing this capability, theory/simulation efforts will address understanding and predicting the structures of these materials based on their sequences, as well as the energy landscapes that define assembly into functional materials. The new cryo-EM suite offers the ability to characterize polymer building blocks in their native, hydrated (or solvated) state (Figure 3.6), including assembled 3D structures via cryogenic electron tomography. Furthermore, isolation of intermediate growth structures and imaging using low keV, high contrast TEM will allow for direct correlation to the energy landscapes inferred by theory.

Nature provides us with a vast library of examples in which hierarchical assembly of nanoscale building blocks (e.g., proteins) generates complex materials/systems exhibiting emergent functions such as self-healing and replication, error correction, and self-regulated ion flow. CINT maintains a core focus on understanding the fundamental principles underlying the assembly of biomolecular components, as well as how interfacial properties control assembly and regulate function. Examples include liquid-liquid phase separation that has recently been shown to regulate transport across the nuclear membrane and likely plays a role in regulating transport in other transmembrane channels and how the nucleotide state of tubulin alters the dimer's conformation in the microtubule lattice, which in turn regulates multiscale assembly and disassembly. Addressing the science in these areas require advancements in both modeling/simulation and characterization. Acquisition of the cryo-CLEM will enable unprecedented characterization of the hypothesized vacancy healing phenomenon that occurs during microtubule annealing. Such data would aid in the further refinement of numerical simulations of microtubule assembly and disassembly dynamics. Understanding of these underlying assembly principles also provides a means of designing new approaches for the hierarchical assembly of synthetic and hybrid materials. Our goal is to exploit biological principles and components to assemble unique materials, such as energy-driven transport for the dynamic assemble large, interconnected networks of lipid and polymer nanotubes.

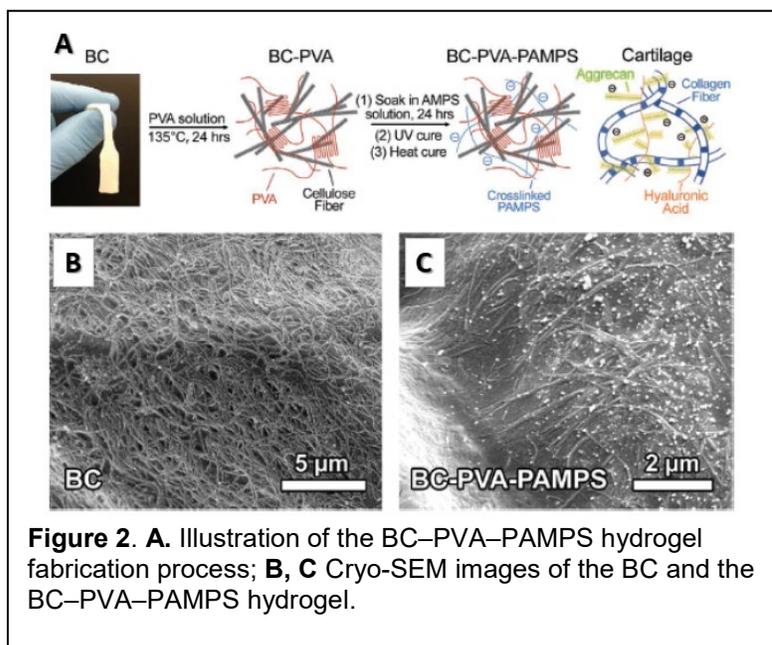
Self-assembly of soft nanomaterials is determined by the interplay among entropy and multiple interactions of similar strengths, leading to many possible equilibrium and nonequilibrium structures. A critical challenge moving forward involves understanding the fundamental physical parameters of soft nanoscale building blocks that give rise to the principles of their self-assembly behaviors. Examples of interest to CINT include polymer-grafted nanostructures, which can assemble into a variety of structures depending on molecular parameters; ion-containing polymers, in which placement and structure of ionic functional groups determine self-assembled morphologies and macroscopic properties; and enzyme-mediated assembly of nanomaterials, where highly selective coupling reactions can direct the assembly of protein hydrogels and enzyme/nanostructural composites. CINT will focus on developing capabilities to determine the design rules for assembly and integration of soft nanomaterials into desired structures. This will

require feedback among synthetic, characterization, and theory/simulation efforts. Simulations on both atomistic and coarse-grained scales provide molecular details of interactions and resultant assemblies. This information will be combined with imaging (cryo-EM and super resolution optical imaging) and scattering experiments to determine mechanisms of assembly, guiding designed synthesis of complex nanostructures.

Addressing these challenges will open the door to materials that display exotic, responsive, and biomimetic behaviors such as light manipulation, self-healing, and adaptivity. As noted above, information-encoded, nanoscale building blocks are a critical component in creating more advanced soft materials. Achieving dynamic and far-from-equilibrium states, however, necessitates innovative approaches that dissipate energy during assembly and organization. These approaches include using external stress or pressure to drive the assembly of nanoparticles into far-from-equilibrium structures by altering the free energy in the system and combining the self-assembly strategy with top-down assembly techniques such as lithography to achieve various non-equilibrium structures.

#### *Advanced optical and electronic imaging of soft matter*

The recently established cryogenic electron microscopy (cryo-EM) lab at CINT houses instruments for the imaging of soft matter, nanomaterials, and beam sensitive materials in their native, hydrated state. Foundational to this lab is a Talos L120C dedicated cryogenic transmission electron microscope (cryo-TEM) for low-dose imaging techniques via user-switchable accelerating voltages (20-120 keV) and a high-speed camera (40 fps at 4k x 4k) optimized for imaging at low keV. A Scios 2 Dual Beam scanning electron microscope with focused ion beam milling capability (SEM/FIB) is also present and optimized for the imaging and sectioning of soft matter and frozen samples. A dedicated cryo-cooled SEM stage from Leica Microsystems provides the capability to FIB section and image frozen samples, allowing for the investigation of buried interfaces. Cryogenic lift-out can be performed on milled samples and transferred to the Talos L120C, thereby covering a wide range of length scales in the characterization of frozen, hydrated samples.



These electron imaging tools are critical for understanding the ultrastructure and interfaces involved in multiscale, hierarchical assembly of soft materials. For example, the cryo-EM suite enabled imaging of a novel hydrogel in its native, hydrated state (Figure 2), which was formed by infiltrating a bacterial cellulose (BC) nanofiber network with a poly(vinyl alcohol) (PVA)–poly(2-

acrylamido-2-methyl-1-propanesulfonic acid sodium salt) (PAMPS) double network hydrogel. Cryo-EM resolved the BC fibers at the surface, while minimizing ice contamination, and demonstrating that BC provided the tensile strength in a manner analogous to collagen in cartilage. In addition, on-going user projects will take advantage of cryo-TEM and cryogenic electron tomography (cryo-ET) to characterize biological assembly across various length scales. For example, cryo-EM experiments will provide real space images of the dynamic 3-D spatial arrangement of the genome (at the chromatin length scale) over time (4D), under different physiologically relevant conditions and following viral infection. The vitrification process is robust to a wide variety of aqueous solution compositions, thus giving us the ability to investigate structural responses to a range of different environmental and epigenetic factors (e.g., cation or viral fragment concentration can be systematically varied). Moreover, by freezing samples at different time points after viral infection, we will be able to probe the dynamic structure of the genome to yield 4D information.

CINT is presently standing up a new capability in cryo-CLEM (cryogenic correlated light and electron microscopy) that will bridge the length scales of optical/fluorescence and electron microscopy into a single process flow that marries the advantages of both techniques. Cryo-CLEM provides the ability to image comparably large field of view using light/fluorescence microscopy to scan broad areas of a specimen and identify specific regions of interest, which can then be imaged with electron microscopy to obtain microstructural information. This technique can be applied to a broad range of nanoscience questions. As noted above, for example, CINT is interested in the self-assembly of microtubule filaments, which involves lattice rearrangement and/or vacancy filling that currently cannot be characterized. CLEM will permit us to visualize and characterize the dynamics of the self-assembly process and locate specific microtubules of interest for ultrastructural analysis via cryo-TEM. This capability will also be used to characterize internal structure and interfaces of soft materials and nanocomposites (e.g., nanoparticle-polymer hydrogels) in their native hydrated state. Here, buried regions of interest can be fluorescently labelled, targeted by fluorescent microscopy and exposed by cryo-FIB. Cryo-ET can then produce 3D tomographic reconstruction of these materials in-situ, combining characterization for target features from the nano- to the micro-scale.

Moving forward, we will adapt the light microscope of the cryo-CLEM capability for super-resolution optical imaging applications. Given the constraints imposed by the cryo-stage and air-immersion objective that allow this microscope to image cryogenically cooled samples, our strategy for improving the signal to noise ratio in the image and increasing the image acquisition rate will be limited to improvements/changes of the light source used to excite fluorescence and the camera(s) used to acquire the images. Structured illumination microscopy (SIM) methods can be used to obtain modest ( $\sim 2x$ ) improvements over the diffraction limit in lateral and axial resolution and provide optical sectioning capabilities. Given the applicability of SIM to almost any fluorescent sample, this approach is often preferable over single-molecule localization microscopy (SMLM) methods that can obtain higher ( $\sim 10x$ ) resolution improvements, but require fluorophores labels that are detectable at the single-molecule level and can be optically switched between emissive and non-emissive states. Both SIM and SMLM techniques can be implemented under the constraints imposed by the Leica cryo-light microscope. Examples of new user science that will be enabled by super-resolution optical CLEM include: (1) studies of the structural basis

of enzyme-substrate interaction (e.g., cellulose-cellobiohydrolase); (2) structural understanding of the hierarchical assembly/disassembly of biological and synthetic filaments; and (3) the use of fluorescent J-aggregates to study of self-assembly.

### 3.4 Nanoscale materials science in extreme environments

#### Scientific opportunity

Our fundamental scientific understanding of how nanoscale features in materials and nanomaterials react to combined external environmental conditions are limited, especially ones that are extreme in nature. Extreme environments exist both in standard operations (turbines in airplanes, electrodes in batteries, and fuel rods in nuclear reactors) and in common materials (electronic materials, additively manufactured materials, and raw metals) when exposed to extreme conditions such as: shock, high-energy irradiation, very-high temperatures, high voltage, high-cycle fatigue, and uncommon chemically reactive conditions. CINT has been a main developer for advanced in-situ characterization and testing of materials at the nanoscale under combined extreme conditions, developing simulation tools to model the materials' responses at the atomic and continuum scales, and in beyond-equilibrium nanomechanical testing. In the coming years, CINT will expand these research efforts into the following areas:

- Nanoscale structure-property relationships of real-materials under extremes
- Unparalleled ion irradiation capabilities to analyze, modify, and synthesize materials under combined environments
- Capturing transients within highly-reactive nanoscale processes

#### Positional science and capabilities

CINT's partnership with the host lab resources, including the Ion Beam Laboratory (IBL, at SNL), the Ion Beam Materials Laboratory (IBML, at LANL), and the Microsystems Engineering, Science and Applications (MESA, at SNL) complex has provided the foundation for our development of new capabilities to offer to our user community. The ion beam facilities have enabled our users to access beam energies ranging from  $100 - 10^8$  eV of almost every element in the periodic table, resulting in hitherto unknown materials being identified in local structure and chemistry. Alteration of a materials' electrical, optical, mechanical, or chemical properties have been afforded via ion implantation; which has been of great interest relative to radiation effects in materials. CINT scientists and their users have demonstrated chemical doping of graphene by selective ion implantation for creating lateral p-n junctions, achieving the smallest photodetector devices in the world. Additionally, these ion irradiation environments may now pair multiple ion beams incident on a sample, to simulate harsh radiation environments in advanced fission/fusion reactors and in space exploration. CINT's greatest impacts have been in the ability to couple these unique ion beam interactions in materials with in-situ imaging, using scanning electron microscopes (SEM) and transmission electron microscopes (TEM). Coupling the ion irradiation environments to other extremes including high temperature, high stress, high



Figure 3.2 Titan ETEM.

strain rate, and fatigue; have added an additional perspective of understanding to the structure-property relationship in nanoscale materials.

Our in-situ ion irradiation transmission electron microscope ( $I^3$ TEM) capability at the CINT-Core enables real-time defect concentration measurement during defect production through concurrent ion irradiation. To determine a material's response to radiation effects, another important parameter besides the defect concentration is the defect mobility during defect production. Recent development of in-situ electrochemical impedance spectroscopy (EIS) at the CINT-Gateway combines ion beam irradiation with an EIS probe to examine both defect content and mobility under irradiation, *in situ*. EIS is a powerful technique to study transport and kinetic processes taking place in a material by measuring the material response to a periodic sinusoidal electrical signal that gives information on the resistance and phase change of the system to external perturbation, such as irradiation. Applying this perturbation across a range of frequencies provides discriminatory and quantitative rate information on how the resistance and phase vary with the time-scale of the thermodynamic, transport, and kinetic processes governing the material response. This new capability could provide unprecedented insight into the behavior of defects as irradiation proceeds, especially those short-lived defects that quickly disappear once the irradiation has stopped. With this new capability, we expect to probe how radiation-induced defects evolve in complex materials relevant for various extreme environments, such as from complex-oxides cladding to potential accident-tolerant ceramic fuels in nuclear energy research and development.

In collaboration with the MESA facility, Discovery Platforms have been developed within CINT and offered externally to the CINT user community to enable in-situ characterization with quantitative property measurements. The Electrochemical TEM Discovery Platform has identified 'hot spots' on electrode surfaces for lithiation of battery anodes (including silicon, aluminum, and gold) with direct correlation between the driven electrochemical profiles and the electrode structural evolution during application of several picoamperes of current. This Discovery Platform has also demonstrated that for high-charge density lithium anodes, contact pressure on the surface of electrodes is required for lithium dendrite suppression. Currently, CINT scientists are working to integrate controlled elevated temperature into this platform, for coupled electrochemical experiments with feedback-verified temperature control. This instrumentation is providing users with the ability to image electrochemical reactions in real-time of customized electrode materials and layouts, which is performing above commercial specifications due to the electrochemical profiles correlating to only the sites being imaged on the electrode.

Improved time resolution has been a focus within CINT for upgrading the current instrumentation for mechanical testing and electron microscopy imaging of ultrafast reactions and mechanisms. One example that pairs these efforts has been on high-cycle fatigue, where CINT scientists monitored fatigue behavior in nanocrystalline copper and platinum by cyclic mechanical loading at  $10^6$  cycles per hour within the  $I^3$ TEM. They found that crack growth occurred at  $\sim 10$  pm per cycle, which broke on average tens of atomic bonds per cycle, which validated indirect observations while refuting age-old metallurgical wisdom. Although not used in this study, CINT's  $I^3$ TEM is capable of ultrafast single-shot imaging for collection of reaction phenomena with either 6 nanosecond or 100s of microsecond frame rates. The other high-performance TEM within the ICNM thrust at CINT is the environmental TEM (ETEM), which allows for gasses and liquid vapors

to be stabilized about a sample within the imaging lens of the microscope without compromising atomic resolution capabilities, where a paired in-situ stage can allow for property measurements within these environments (Figure 3.7). The ETEM has been outfitted with a high-speed direct electron detection camera, allowing for imaging up to 1,500 frames per second (fps), greatly increasing temporal resolution of various in-situ reactions beyond video frame rates (30 fps). The increased temporal resolution paired with our expansive in-situ environmental capabilities brings CINT to the forefront for mechanistic understanding of structure-property relationships in extreme environments for nanoscale materials.

### Toward the future

#### *Nanoscale structure-property relationships of real-materials under extremes*

To understand how properties of nanomaterials change when exposed to harsh environments, including matrixed combinations of ion irradiation, high temperatures, high cycling, chemically reactive, and high strain rates. CINT's collection of advanced in-situ characterization techniques combined with new Discovery Platforms in collaboration with MESA, will provide first-of-their-kind testing platforms for real-materials integration. Two TEM compatible Discovery Platforms identified are the Electro-Thermal Discovery Platform for 2D materials (Figure 3.8) and the Mechanical-Environmental-Thermal Discovery Platform which can test samples within liquid-environments. Each of these designs are manufactured to enable users to access their quantitative property measurements during nanoscale imaging within a TEM, with plans for platforms to be available for transport to user's home institutions for use. The coupled property measurements on a single platform enable more versatility in the environments for testing, especially when these MEMS platforms are paired with the environmental SEM, ETEM, or I<sup>3</sup>TEM. CINT will use these tools *to determine what the fundamental, atomic-scale processes are that describe the response of materials to coupled extreme environments, e.g. irradiation and corrosion*. Simulations of these environmental structure-property relationships are being formulated to compare to the experimentally generated data, where these modeling capabilities will become available to users through the Virtual Electron Microscopy Laboratory.

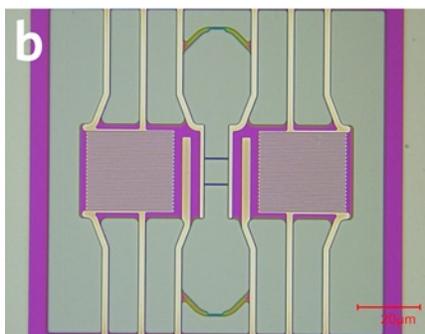


Figure 3.3 Prototype of Electro-Thermal Discovery Platform fabricated in the CINT Integration Laboratory.

#### *Unparalleled ion irradiation capabilities to analyze, modify, and synthesize materials under combined environments*

Planned upgrades being implemented at both the IBML and IBL will enable unparalleled ion irradiation capabilities combined with extreme environments. Dual-beam irradiation capabilities to enable in-situ reactions of neutron displacement damage by self-ions and transmuted impurity effects by He ions will enable users to study reactions environments like those within the core of advanced nuclear reactors. Coupling a liner plasma device to the dual-beam irradiation environment will provide unmatched capability in the world to study plasma material interactions in the core of magnetic confinement fusion devices. In this environment, the plasma facing material components are exposed to unprecedented conditions of very-high fluxes of low energy fuel particles, extreme thermal loads, and fast neutrons. The development of positron annihilation spectroscopies (PAS, both lifetime spectroscopy and coincident Doppler-broadening

spectroscopy) at the CINT Gateway will be used to investigate small vacancy clusters and void formation and evolution dynamics in solids during ion irradiation. This in-situ PAS capability will be highly sensitive to initial vacancy generation and small cluster formation behavior (too small to be seen by TEM), critically needed experimental data to compare directly with advanced modeling and simulation results. Additionally, an increased ion energy range is planned at the IBL, for beams up to 100 MeV for Au ions, and as low as 100 eV for light elements. These ion beam lines are already compatible with an array of characterization tools, including the I<sup>3</sup>TEM and the in-situ ion irradiation scanning electron microscope (I<sup>3</sup>SEM), which affords additional in-situ environmental control in temperature, strain, electrical, gaseous, and liquid environments. Materials synthesis using ion irradiation is also being developed, especially in the use of controlled environments to direct the location of defects and impurities to obtain unique properties in nanoscale materials. These include physical vapor deposition with concurrent surface modification/perturbation by low energy ion bombardment; non-equilibrium chemical doping under elevated temperatures by direct ion implantation; and liquid-based synthesis in the presence of highly charged electronic states and radiolytic species as a result of high energy ion energy deposition. In particular, the Nanoimplanter at the IBL and ultra-low energy Colutron ion beam system at the IBML (50 eV – 10 keV) are developed to perform deterministic defect doping with nanoscale resolution, and 2D material doping in optoelectronics and quantum materials research. Improvements in our simulations are being pursued to reflect these planned upgrades for in-situ experiments and synthesis routes using ion irradiation and implantation capabilities.

#### *Capturing transients within highly-reactive nanoscale processes*

Increasing the temporal resolution of our current instrumentation, to a level that can match simulations will enable transients within highly reactive nanoscale processes to be captured. A Relativity System and a direct electron camera on the I<sup>3</sup>TEM improves the frame rate of the camera by over 100x, for all in-situ and ion irradiation experiments performed on this tool to greatly expand the temporal resolution of nanoscale mechanisms. *We will use this advancement to characterize the transient states in nanophenomena to better understand their mechanisms, and we will use simulations to verify the generated images and diffraction patterns.*

Our emphasis is on coupled-property measurements and enhanced in-situ characterization, towards operando analysis at conditions not being pursued at other research institutions. The new capabilities being developed will allow for the exploration of understanding extreme complex mechanisms in nanomaterial systems including new capabilities for ion beam analysis, modification and synthesis, and transient-state understanding of nanomaterial processes. These ideas showcase the collective interest in the fundamental understanding of unexplored nanoscale phenomena under extreme in-situ conditions. Our vision is aligned with the call in the DOE Basic Research Needs report on the ‘Future of Nuclear Energy’ and ‘Transformative Manufacturing’, and the question posed in the ‘Challenges at the Frontiers of Matter and Energy’ BESAC report, *“How do we characterize and control matter away – especially very far away from equilibrium?”*

### 3.5 Machine learning and artificial intelligence in nanoscience

#### Science opportunity

Machine learning (ML) has been around since the dawn of digital computing. But recent advances in computing hardware and algorithm development, along with an exponential acceleration in the quantity of scientific data being collected, have triggered the proliferation of machine learning throughout all fields of science. In the rush to utilize these new tools, there is still much to learn regarding how to apply these methods in a way that advances our fundamental understanding of the physical world, and in our specific case the understanding of nanoscale phenomena. The most compelling applications of machine learning will be toward incredibly difficult problems and where the outcomes are not known *a priori*, such as materials subjected to extreme environments, autonomous optimization of synthetic routes for new materials discovery, and intelligent/autonomous data collection. The largest impacts will be from data analysis that is not tenable by human processing, either because of the needed response time or the enormous quantity of data.

#### Positional science and capabilities

CINT has several well-developed efforts utilizing machine learning and artificial intelligence, and has partnered with collaborators at academic institutions who are recognized leaders in this area. For example, we have developed a strong partnership with Professor Rajiv Kalia, at the University of Southern California, and have started collaborations with several other USC professors from within the Information Sciences Institute (ISI). CINT is also taking advantage of enduring relationships with Sandia's academic alliance partners as a source for students, postdocs, and collaborators with expertise in ML/AI. These partnerships allow us to leverage existing external capabilities and expertise to address novel nanoscale challenges.

Our current ML expertise and capabilities include automated qubit optimization, prediction of new synthesis routes (physical vapor deposition) for novel metastable nanostructured alloys, virtual microscopy, and acceleration of quantum simulations. Building upon these capabilities will allow us to make scientific advances in areas of fundamental importance to CINT, such as quantum materials, materials response to extreme environments, and nanomaterials discovery.

#### *Automated qubit optimization*

Semiconductor qubit devices are becoming more complex as more qubits are added to 1D arrays and further extended into 2D qubit arrays. As the number of qubits increases, so do the number of controls and cross-correlations between controls. Configuring these complex devices for qubit operation quickly becomes intractable for human experimenters. CINT has developed the capability to autonomously tune qubits using ML and image analysis techniques. This capability, which will continue to evolve, will accelerate scientific discovery through full, autonomous, device configuration.

#### *Predicted synthesis of metastable phases*

CINT has developed a reduced-order model for accelerated microstructure evolution predictions by utilizing time-series multivariate regression splines or long short-term memory (LSTM) deep learning algorithms within a phase field simulation. We demonstrated this capability to predict microstructure evolution during spinodal decomposition and also are applying it to mesoscale models for corrosion (atmospheric corrosion and molten salt corrosion). This same framework

will be used to predict synthesis routes (physical vapor deposition) for novel metastable nanostructure alloys with properties desirable for combined extreme environment applications.

#### *Accelerating quantum simulations*

Quantum chemical computation is a foundation of materials science but is limited in application due to well-known issues of high computational cost, small system sizes, and short timescales. Emerging data science approaches promise to break the existing scaling barriers and enable training of neural networks capable of quantitative predictions for much larger materials system as well as reducing the cost of these calculations to a level similar to classical force fields. CINT has already demonstrated this capability using a transfer learning algorithm, trained on 5 million highly accurate density functional theory calculations, to generate a coupled cluster model that retains quantitative accuracy. The resulting models will encode chemical and physical information that is extensible to much larger systems, and transferable to new types of processes.

#### *Toward the future*

As the application of ML toward nanoscience is evolving at an amazing pace, so will our strategy. CINT will naturally focus our ML efforts in areas where we are unique and leading the community, such as quantum information science, nanomaterials Discovery Platforms, and the response of materials to extreme environments.

#### *ML for quantum information science*

Semiconductor quantum dots are a promising platform for quantum computation due to their nano-meter scale footprint per-qubit. However, as previously mentioned, configuring semiconductor quantum dot devices into the low electron regime can be intractable for a human controller, particularly as the number of quantum dots in a device increase. It has already been demonstrated that image recognition and automatic device control can increase the configuration time by an order of magnitude when ‘fine tuning’ a partially configured device. CINT will continue supporting and advancing this field by: (1) providing software tools for perception, control, and intelligent agents to control semiconductor qubit devices and (2) providing experimental capabilities, test devices, and expert experimental guidance for users to test their ML and automation routines.

#### *ML for synthesis of nanomaterials*

Nanoparticle synthesis is a main underlying aspect of nanoscience. Understanding and controlling synthesis would have profound impact by enabling new experiments similar to the advances in biology that were enabled by the ability to precisely synthesize proteins. We are developing ML systems for controlling nanoparticle reaction platforms. These systems will learn from a wide range of real time diagnostic data and inputs. The output will be optimal control parameters to achieve the desired synthesis. This will prove especially useful for applications such as magnetic nanoparticles where size uniformity and repeatability are crucial. Beyond nanoparticles, CINT will pursue using machine-driven routes for discovering routes to synthesize novel nanostructured alloys for resilience against combined extreme environments.

#### *ML for materials in combined extreme environments*

The main challenge in the current modus operandi of the characterization, theory, and simulation of materials degradation mechanisms in extremes (high strain rate, radiation, high temperature,

corrosion) is to develop both data-driven capabilities and modeling paradigms that can integrate meaningful data in order to interrogate and probe compositional and structural changes simultaneously at appropriate spatial and temporal resolutions. At least three significant gaps contribute to this corollary: the first gap is the lack of meaningful descriptors and knowledge of structure-property relationships to derive insights from abundant data. The second gap pertains to the fact that most of the machine learning methods applied to date, both theoretical and experimental, are relatively straightforward adaptations of methods originally developed for other problems, such as image recognition for example. And the third gap is the lack of approaches for effective fusion of data gathered from experiments and simulations.

To address these gaps, we are developing a virtual operando microscopy capability, which involves using ML for fingerprinting complex mechanisms as they evolve, optimizing functionalities of nanostructured materials, and classification of defect structures and their signatures in large scale molecular simulations. We are also focusing our efforts on the ability to directly compare diffraction patterns and transmission electron microscopy images from atomistic simulations and experimental efforts. Coupling atomistic simulations with TEM and energy-dispersive X-ray spectroscopy (EDS) provides a means to understand defect structures and densities via comparison of arrays of diffraction patterns (virtual diffraction) with nanoscale resolution extracted from atomistic simulations of the same geometries/chemistries.

### 3.6 Scientific expertise to realize our vision

Nanomaterials integration involves: (1) Synthesizing and fabricating individual nanoscale building blocks, which may be combined to form specific heterostructures, (2) characterizing their functionalities, (3) understanding and predicting their fundamental chemistry and physics, (4) assembling these building blocks, and (5) delivering a functional material system. The fundamental challenges underlying such integration go beyond complex fabrication or the engineering of known solutions; they lead to novel discoveries and new sciences.

Currently, the four thrusts as described in Section 2 (Quantum Materials Systems, Nanophotonics and Optical Nanomaterials; In-situ Characterization and Nanomechanics; and, Soft, Biological, and Composite Nanomaterials) have already demonstrated their specific strengths and scientific expertise at the international level. A wide variety of activities is being initiated within the four thrusts and our user community, including research activities, user projects, new instrumentation, Discovery Platforms, and cross-thrust activities. These areas are being developed to be mutually supportive and to maximize their value to our user program and the scientific community. Our internationally recognized expertise in different scientific and technological fields has positioned ourselves in a leadership role in the development of novel capability in supporting users and emerging nanoscience research.

However, as both nanoscience programs and nanomaterials integration continue to evolve in response to new scientific challenges in the scientific community, we will need to not only retain our current workforce to remain in the forefront of nanomaterials integration, but also to strategically hire more scientists and technologists with expertise in fields such as system level modeling, architecture designs, and in-situ multi-length scale/temporal characterization of materials to address the dynamic integration challenges and other challenges described in Section 3 to realize CINT's overall vision of nanoscience.

With our current and expanded expertise, we will maintain our leadership role in nanomaterials integration. Our scientists will continue to actively engage our user community through a variety of mechanisms to promote the integration of nanomaterials and to address new scientific challenges, through both world-leading research and the development of unique capabilities important for the future of nanoscience and nano-manufacturing.

## 4 FOUNDATIONAL AND TRANSFORMATIONAL CINT CAPABILITIES

Through a combination of research expertise, specialized capabilities, and essential foundational techniques, CINT enables our users to perform multidisciplinary research that would otherwise not be possible. Therefore, to address the research challenges of the future, CINT is continuously improving its current capabilities, exploring concepts that can lead to new experimental, theoretical and computational methods, and selectively disinvesting in capabilities that are no longer state-of-the-art (Figure 4.1).

In this section, we identify a selection of CINT capabilities that will be important in addressing future integration challenges and that will transform the way integration research is conducted (CINT strategic objective 2). Some involve significant upgrades to current capabilities while others will require entirely new efforts that build on our expertise.

In addition to the specialized capabilities highlighted here, CINT has many more which form an essential foundation for the majority of nanoscience research. The complete list can be viewed on our website.<sup>4</sup> As specified in CINT strategic objective 4, CINT must continue to innovate this infrastructure to have the baseline instruments that all researchers need to properly measure and control the relevant conditions in their experiments.

By virtue of having two DOE host laboratories, CINT is uniquely able to leverage a wide range of expertise plus substantial prior investment in research infrastructure. While there are several examples of this synergistic benefit in our capabilities, perhaps the most differentiating among the DOE Office of Science user facilities is our portfolio of Discovery Platforms.



*Figure 4.1 Addressing integration through collaborative capabilities and expertise in fabrication, synthesis, characterization, and theory.*

### 4.1 Discovery Platforms: A CINT signature initiative

The need to reproducibly characterize individual nanostructures or synthesize nanomaterials with exquisite chemical control has inspired CINT to develop Discovery Platforms. These unique research tools consist of micro-fabricated structures or devices for specific nanoscience experiments. Simple platforms are fabricated in CINT whereas the most complicated devices, essentially a lab-on-a-chip, are designed and produced in cooperation with SNL's Microsystems Engineering Science and Applications (MESA) facility.

Discovery Platforms are conceived, designed, and commissioned with user input and active participation. They allow users to conduct experiments that are not possible using traditional laboratory equipment.

Responses of the next-generation atomically thin materials exhibit strong dependence on strain and temperature, yet currently no tools or methods exist to probe strain- and temperature-coupled electronic transport at the atomic level. This significantly limits our ability to identify the

<sup>4</sup> <https://cint.lanl.gov/>

dominant scattering mechanisms and develop new approaches for enhancing charge mobility in these materials. CINT is developing two unique *Electro-Thermal and Electro-Mechanical Discovery Platforms*, that enable users to apply thermal and strain gradients, respectively, to atomically thin materials while simultaneously measuring their electrical and thermal transport properties. These first-of-a-kind multi-purpose platforms are compatible with nanoscale in-situ characterization in the SEM, TEM, X-ray, and scanning probe microscopies, and provide users with enhanced dynamic control over the properties of individual nanomaterials, both 1D (nanowires, carbon nanotubes) and 2D materials. The platform will be used to study broadly defined structure-electrothermal property relationships in nanomaterials.

The *Electrochemical Discovery Platform* is a microfabricated fluidic platform designed to study electrochemical energy storage processes in real time inside a transmission electron microscope, as shown in Figure 4.2. This platform enables direct observations of solid/liquid interfacial processes, such as electrode/electrolyte interactions, electrode dissolution in electrolyte, and solid-electrolyte interphase layer formation.

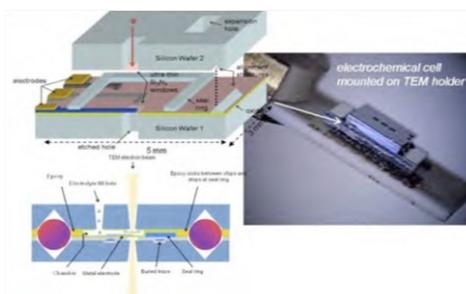


Figure 4.2 CINT's Electrochemical Discovery Platform.

The *Microfluidic Synthesis Discovery Platform* is an extremely flexible system for nanoparticle synthesis, functionalization, and real-time characterization. The microfluidic system utilizes an array of glass or plastic chips with channels that can hold volumes from 50 microliters to one milliliter. Design features include droplet forming junctions, mixing segments, and serpentine channels all in a single form factor with a standardized chip header for fluidic connections and precision temperature control. Real-time reaction monitoring via visible and fluorescence microscopy, as well as UV-visible and near-IR spectroscopy are currently being used with mid-IR spectroscopy and dynamic light scattering to be added in the future. A central issue in the development of multi-scale materials is large-scale predictive assembly of nanoscale building blocks (e.g., colloidal nanoparticles, polymers, proteins, etc.) into functional mesoscale assemblies. One approach to address this limitation is the development of experimental systems that can achieve reproducible synthesis and most importantly, assembly of the building blocks. Work in this area will build on CINT's *Microfluidic Discovery Platform* and expand into more complex architectures that enable the rapid prototyping of molecular building blocks, as well as the controlled assembly of soft matter, particularly into 3D mesoscale assemblies. In a new development, there will be a strong emphasis on integrating the *Microfluidics Discovery Platform* with new capabilities and techniques developed as part of the in-situ characterization effort, as described above. The key objective will be developing new Discovery Platforms that enable tight coupling of in-situ characterization, ML/AI, and microfluidic delivery, enabling real-time control over multi-scale, hierarchical assembly.

In the area of quantum information sciences, CINT is focusing on development of the first-of-its-kind *QSNMR Discovery Platform*. The QSNMR platform will rely on qubits' extreme sensitivity to magnetic fields to perform magnetometry and NMR spectroscopy at the smallest scale possible: a single spin. This new project will cover three areas: (1) precisely fabricating magnetometry-

ready NV centers in nanodiamond using focused ion beam implantation, (2) quantum sensing of magnetism and NMR using NV centers, and (3) demonstration of *QSNMR Discovery Platform* on “exemplar” systems of semiconductor QD and designer quantum materials. Ultimately, this work will set the stage for a new future in nanoscience research, because, once this measurement system is calibrated and proven, it will serve as an infrastructure for user scientists in multiple fields to measure atomically precise, high-resolution magnetic properties.

## 4.2 Synthesis and fabrication

Creating novel and integrated nanomaterials requires robust methods to reproducibly synthesize nanoscale constituents with known composition and structure, rigorous control of assembly processes to organize the components (“bottom-up assembly”), and microfabrication tools (“top-down fabrication”) to prepare architectures that connect nano-to-macro length scales. CINT currently offers and continuously develops these state-of-the-art capabilities including the following.

### III-V Semiconductor molecular beam epitaxy (MBE)

The two CINT systems are in demand worldwide to grow high-purity, ultra-high mobility AlGaAs-based III-V compound semiconductor structures with atomic monolayer precision for fundamental studies of 1D and 2D nanomaterials, as well as nanophotonic materials, including quantum cascade lasers and quantum well infrared photodetectors. Our future plan includes the addition of Indium to both machines to allow the growth of strained In-alloys on GaAs substrates and strained In-alloys on InP substrates.

### III-V Semiconductor Metal Organic Chemical Vapor Deposition (MOCVD)

This capability, Metal Organic Chemical Vapor Deposition (MOCVD), enables the growth of complex nanostructures based on the III-V and III-nitride (AlGaInN) semiconductor materials systems, including nanowires (NWs) and quantum dots (QDs). Capabilities are available for the preparation of structures involving diverse semiconductor families such as large band gap semiconductors (i.e., III-Nitrides, boron nitride (BN)) and low band gap materials (e.g., InGaAs/InP, III-antimonides). Traditionally, nanomaterial system fabrication has been carried out in planar structures. More recently, however, 3D architectures have started to emerge in micro and nano-electronics that will ultimately expand their fundamental properties to unique phenomena. Current epitaxial capabilities available to CINT users include the following: four III-Nitride MOCVD systems; three commercial and one home built high temperature systems; two III-V (non-nitride) MOCVD systems; two MBE III-As systems; and two MBE III-V systems.

### Heteroepitaxial growth

Nanocomposite films with specific electrical, optical, magnetic, thermal, and/or superconducting properties are grown by pulsed laser deposition (PLD), polymer assisted deposition, and laser MBE. Upgrades in progress will enable us to grow epitaxial nanocomposite films incorporating nanoparticles of controlled particle sizes and densities at desired locations.

### Quantum materials synthesis

CINT has synthesis capabilities for 2D TMDCs and topological insulator (TI) thin films via solid source chemical vapor deposition (CVD) and pulsed laser deposition (PLD). Future directions are heterostructures composed of quantum materials for novel functionalities and scaling up with

precise control of physical characteristics of 2D TMDCs and TIs. Metalorganic CVD and physical vapor deposition with an in-situ monitoring system of quantum materials growth will be developed.

### Nanowires

CINT specializes in the synthesis of semiconductor nanowires by solution-phase, CVD, and physical vapor deposition (PVD) approaches to produce single crystal nanowires, radial/axial heterostructured nanowires, and complex architectures consisting of Si/Ge and compound (II-VI and III-V) semiconductor materials. Future directions include advances in atomic-level control to introduce embedded quantum dots and quantum wells, for example.

### Atomic-precision fabrication

One of our newest capabilities enables placement of dopant atoms into silicon with atomic-scale precision by using hydrogen resist lithography combined with scanning tunneling microscopy. This technique yields the ultimate in atom-by-atom construction of nano-electronic devices. CINT is one of only a few institutions in the world with this capability and the only user facility to offer the capability (Figure 4.3).



*Figure 4.3 Atomic-precision fabrication capability.*

### Integration laboratory

This fabrication laboratory is a class 1000 cleanroom with a comprehensive suite of micro/nanoscale tools including atomic layer deposition (ALD), low pressure CVD, PVD, electron beam and photo lithography, and focused ion beam. Future capability enhancements include: the creation of a soft lithography capability to enable users to fabricate and integrate soft/hard nano/microstructures; expansion of deposition/etching tools to meet user demand; addition of noble metal ALD; and, replacement of legacy tools with the current state-of-the-art in commercial instrumentation.

### Solution phase synthesis of optical nanomaterials

CINT is pioneering new optical nanomaterials with advanced functionality and the methods used to create them. We have developed a suite of novel photostable quantum dots that afford non-blinking and non-photobleaching emission from the visible to the infrared. Like other colloidal nanoparticles, these quantum dots are solution processible and can be used as “inks” in the production of thin films, polymer composites, or integrated devices. Their enhanced stability affords chemical robustness and the ability to perform otherwise inaccessible experiments. CINT also possesses a fully automated batch reactor system (FABRS) and a microfluidic setup for substrate-supported synthesis in-flow. These custom synthesis tools enable multistep (>100) reactions to be conducted under software control and solution-phase nanowire growth akin to a CVD process, respectively. Together, the unique methods support new materials discovery and optimization, as well as rapid data acquisition and a pathway toward in-situ monitoring and control of reactions ‘on-the-fly.’

### Chemical vapor deposition capability for growth of semiconducting nanowires and thin films

Our 3-inch wafer-scale cold wall CVD reactor allows preparation of high-quality and electrically-doped nanowire and thin film heterostructures with well-controlled axial or radial interfaces. We

offer a unique capability for flowing metal-organic precursors that provides fine control over elemental composition of catalyst seed particles and abrupt interface generation. This capability is thus available for generation of a wide range of material types, compositions and architectures (including Si/Ge, III-V, and other compound semiconductor materials), for exploration of light emission, energy harvesting, energy storage, and optoelectronics applications.

#### Carbon nanotube processing and functional modification

Development of carbon nanotube-based materials is enabled by CINT's processing capability in both aqueous and organic media. Nanotube samples may be provided as surfactant or polymer suspensions and in sol-gel, aerogel, and polymer matrices. We use state-of-the-art density gradient and aqueous two-phase processing approaches to separations of semiconducting and metallic tubes and by single chirality with expertise for generating filled and empty tubes of variable length. Non-covalent functionalization methods are providing systems for fundamental surface chemistry studies and for the understanding of separations mechanisms. We are pioneering low-level covalent doping strategies via solution and solid-state methods for the introduction of new photoluminescent emitting states of interest for enhancing quantum yields and introducing new functionality.

#### Fabrication of metamaterials and plasmonic nanostructures

World-class tools for lithographic fabrication to sub-10 nm length scales and epitaxial growth form the core of CINT's world-leading creation of innovative concepts in nanoresonator/nanoantenna, metamaterial and metasurface for manipulation of light. Fabrication with active and passive metallic, semiconductor, dielectric, complex multiferroic, and emerging 2D materials can all be met to generate hyperbolic metamaterials and arrays of meta-atom structures on few-layer heterostructures. Realized functionality includes anomalous refraction, flat optics, customized thermal emission, wavefront engineering, and active control of response such as polarization and phase, and the ability for light concentration and spectral tuning. Light manipulation can be attained from THz to near-IR. Fabrication efforts are strongly supported by off-the-shelf simulation packages running on high-performance workstations and a unique set of CINT-developed capabilities in functional nanomaterial integration including integration with semiconductor heterostructures grown by high quality molecular beam epitaxy.

#### Dip-pen nanolithography (DPN) and custom AFM-nanopipette-optical microscopy system for nanointegration and nanofabrication

As a scanning probe lithographic technique, the CINT DPN capability is highly complementary to other lithographic approaches for materials integration and heterostructure generation. We have advanced the tool for delivery of liquid inks onto any desired substrate with precise positioning in the nanometer range. Our focus is on extending DPN to "large" (10-50 nm diameter) nanocrystals mixed into simple solvent carriers, with an ability to write controlled numbers of nanocrystals—from single to monolayer spots or lines—onto 1D, 2D, and 3D pre-patterned structures of both soft and hard materials. Integration of optical emitters to dielectric optical antennae, plasmonic structures, and metamaterial assemblies has been demonstrated (Figure 4.4). A new dual scanning probe microscope for nanomaterial and chemical reactant delivery, outfitted with an optical microscope, will enable real-time optical characterization of integrated systems and optical defects "chemically implanted" in a substrate, e.g., 2D TMD.

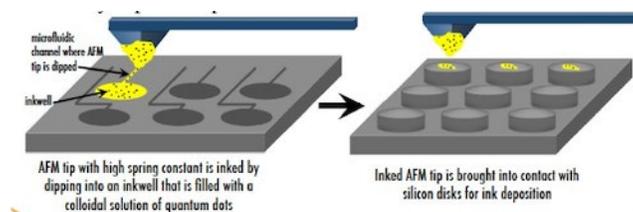


Figure 4.4 DPN using a quantum dot ink.

### Flow-reactor synthesis of nanoparticles

A computer controlled microfluidic synthesis platform that is capable of both systematic variation of synthesis and continuous synthesis without variation. The system features real time spectroscopic detection of nanomaterial properties to provide feedback and control of materials synthesis.

### High-throughput synthesis and evaporation platform with *in situ* optical characterization

Advanced heating and stirring modules have been assembled to run up to 36 larger (up to 15 mL) or 78 smaller (up to 3 mL) reactions, e.g., metal organic framework (MOF) synthesis or supercrystal self-assembly, simultaneously, with controlled evaporation option. The new system will enable harnessing multiscale control over framework molecular synthesis, composite formation, and supercrystal assembly. It affords precise control over convective mixing, providing an ultrastable temperature profile, as well as sedimentation processes, allowing a new degree of size, facet and morphology optimization. *In situ* / real-time characterization is provided by UV-Vis-SWIR absorption and photoluminescence probes.

### Expression and purification of engineered functional biomolecules

A library of engineered biomolecular machines to enable their integration and control for the fabrication of hybrid nanomaterials and systems. As an example, kinesin motors that contain a unique ion-binding site that may be used as a molecular on/off switch for motor function.

### Amphiphilic monomers/polymers for preparation of durable artificial membranes

These techniques can be used to prepare polymersomes, supported lipid bilayers, and genetically encoded polymers. Custom synthesis of monomers that can be self-assembled into organized mesophases and captured into durable yet stimuli-responsive polymer networks. These networks are hierarchically ordered and can be used to order nanoparticles across multiple dimensions.

### Physical Synthesis of Nanostructured Materials

The physical vapor deposition (PVD) system is used to synthesize nanostructured metal, alloy, ceramic, or composite materials with controlled layer thickness, grain size, or particle size. Energetic ion or neutral atom bombardment during growth can be used to tailor the nanostructuring dimension, residual stress, texture, epitaxy, etc. Post-deposition vacuum annealing or ion-bombardment facilities are also available for modification of the PVD-synthesized materials.

### 4.3 Characterization

CINT's current characterization capabilities include an extensive array of differentiating techniques to determine structural coordinates with atomic resolution, chemical composition, and temporal behavior for individual nanostructures to complex integrated environments. Our future emphasis will be to develop methods to extract such information from increasingly complex hierarchical materials with nanoscale to mesoscale complexity. The development of advanced probes plays a critical role in high-impact nanoscience discoveries and innovation of next generation technologies.

#### Transmission electron microscopy (TEM)

The capabilities at CINT offer an array of in-situ techniques for correlating dynamic structural information with associated electrical, mechanical or compositional changes. CINT Discovery Platforms are routinely employed as in-situ TEM sample holders to provide unprecedented fundamental information on liquid/solid interfacial reactions, individual nanowires, and related structures. CINT has recently expanded its TEM capabilities with the addition of an image corrected environmental TEM with a Gatan K2 - single electron camera. This camera greatly increases the potential systems that can be investigated using CINT's Discovery Platforms by minimizing beam damage by up to two orders of magnitude. CINT future plans for TEM include: significantly expanding our capacity to meet the consistent demand for instrument time by highly-ranked user proposals, and developing the expertise and techniques for soft and composite nanomaterials microscopy needed by the integration challenges in Section 3.

#### In-Situ Ion Irradiation Transmission Electron Microscopy Facility (I<sup>3</sup>TEM)

The I<sup>3</sup>TEM facility combines a 200 kV JEOL 2100 high-contrast TEM (2.5 Å point resolution) with a 10 kV Colutron and a 6 MV Tandem accelerator. The I<sup>3</sup>TEM facility can permit a wide breadth of combined experiments in high temperature, flowing liquid, gas exposure, mechanical loading, displacement damage, gas implantation, and numerous sequential or simultaneous combinations thereof to evaluate the structural evolution that occurs during ion beam modification or overlapping combinations of extreme environments.

#### In-Situ Ion Irradiation Scanning Electron Microscopy Facility (I<sup>3</sup>SEM)

The I<sup>3</sup>SEM facility combines a 30 kV JEOL IT300HRLV SEM (1.5 nm spot resolution) with a 6 MV Tandem accelerator. The I<sup>3</sup>SEM facility can permit a wide breadth of combined experiments in high temperature, gas exposure, mechanical loading, displacement damage, gas implantation, and numerous sequential or simultaneous combinations thereof to evaluate the structural evolution that occurs during ion beam modification or overlapping combinations of extreme environments. Specific capabilities of the I<sup>3</sup>SEM include:

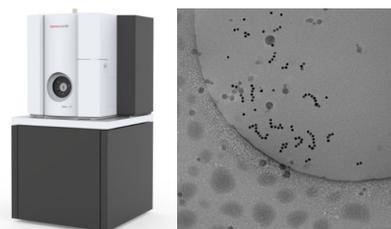
- Three in-situ SEM mechanical loading frames: Hysitron PI-85 nanoindenter, MTI/Fulham heat straining stage, custom-built fatigue stages.
- Large variety of ion species in the range between protons and gold.
- Ion beam currents from single ion strikes up to 100 nA (Tandem).
- Electron beam or ion beam induced current measurement (EBIC or IBIC).

### Liquid Cell Discovery Platform

The CINT Liquid Cell Discovery Platform is an in-situ TEM sample holder that allows for quantitative measurements of nanoscale electrochemistry and nanoscale chemical reactions with an integrated lithographically defined heater. Future plans for this platform include adding a mechanical actuation stage.

### Cryogenic electron microscopy (Cryo-EM)

The recently established cryogenic electron microscopy (cryo-EM) lab at CINT houses instruments for the imaging of soft matter, nanomaterials, and beam sensitive materials in their native, hydrated state. Included is a Talos L120C dedicated cryogenic transmission electron microscope (cryo-TEM) from Thermo Fisher Scientific (Figure 4.5). The cryo-TEM has low-dose imaging techniques, user-switchable accelerating voltages (20-120 keV) and a high-speed camera (40 fps at 4k x 4k) optimized for imaging at low keV. It has a modified pole piece with a larger gap which allows for a wider angle of rotation for tomography experiments. It also accepts the insertion of a 'cryo-box', which gives greater stability when imaging cryogenically frozen samples, and also ensures that samples are kept in a cryogenically cooled environment from preparation, right through to imaging. Also present is a Thermo Fisher Scientific Scios 2 Dual Beam scanning electron microscope with focused ion beam milling capability (SEM/FIB) optimized for the imaging and sectioning of soft matter and frozen samples. A specialized in-column T3 detector is included, which is optimized for low-kV, low energy secondary electron detection imaging. A dedicated cryo-cooled SEM stage from Leica Microsystems provides the capability to FIB section and image frozen samples, allowing for the investigation of buried interfaces. The Leica cryo-workflow is specially designed to allow sample preparation and transfer while maintaining a frozen state and includes cryogenically cooled shuttle (VCT500), sample transfer workstation (VCT) and sputter coater/freeze fracture unit (ACE600). Cryogenic lift-out can therefore be performed on milled samples and transferred to the Talos L120C, thereby covering a wide range of length scales in the characterization of frozen, hydrated samples. High sensitivity electron backscatter diffraction (EBSD) and energy dispersive X-ray spectroscopy (XEDS) cameras are also attached. An additional glove-box attachment for the VCT will soon be installed, allowing samples to be loaded onto the cryo stage directly from an inert atmosphere. Finally, CINT will be adding the instrumentation to perform cryo-CLEM (cryogenic correlated light and electron microscopy).



*Figure 4.5 CINT's cryo-EM capability investigates soft matter and nanomaterials in their native, hydrated states.*

### Nanomechanics

CINT is currently the only NSRC with a dedicated nanomechanics capability, including high-throughput statistical measurements at the nanoscale under ambient or multiple extreme environments (e.g. temperature, radiation, fatigue, shock) in combination with dynamic observation of mechanisms using a variety of in-situ TEM and SEM test platforms. Beyond modulus and strength, the CINT tools can be used to explore a broad range of relevant physical processes from tribological degradation to interfacial delamination. Nanoscale measurements are complemented with nano- to macro-scale capabilities for an integrated multi-scale experimental suite. Tools include several nanoindenters, SEM/TEM-based loading platforms, and

the Bruker Dimension Icon Quantitative Nanomechanical module, which is also indentation capable.

#### Diamond nitrogen-vacancy magnetometry

We have established a nitrogen vacancy (NV) quantum sensing capability at CINT. DC magnetic fields of nanomaterials can be measured at room temperature using state of the art microwave and optical control enabling nuclear magnetic resonance and electron spin resonance experiments at the nanometer scale. We have also assembled a “quantum diamond magnetometer” for DC wide field imaging which enables DC magnetometry over a 2x2mm field of view with 500nm resolution.

#### Ion implant with nanoscale patterning

Nanoscale ion implantation is a capability for low energy, high resolution focused ion beam implantation. This instrument is equipped with a mass velocity filter allowing implants of a wide range of atomic species, in-situ electrical probes, a high-resolution laser interferometer driven stage and a dual beam SEM. Available atomic species include Si, Au, Ni, Cu, P, Ga, Sb, Pt, Er, Ge, V, Cr, Fe, Zn, Sn and Te.

#### Quantum transport

The quantum transport capability includes multiple instruments configured for measuring low temperature electrical transport in semiconductor devices. We have demonstrated single shot electron spin measurements and rapid pulsing of the semiconductor nanostructure energy levels using these advanced techniques.

#### Surface sensitive probe

The low energy electron microscope (LEEM) is a unique and versatile surface microscope that can be used to view dynamic processes on surfaces in real time with a spatial resolution of 7-8 nm and a depth resolution of one atomic layer. We extend this capability to conduct photoemission electron microscopy (PEEM) by the addition of various UV sources to enable probing of the electronic structures of surfaces and nanomaterials. Future developments include the addition of deep-UV CW lasers, which will support high lateral resolution (10-20 nm) electronic structure mapping and magnetic imaging of surfaces and nanomaterials, and an in-operando microscopy capability with electrical biasing.

#### Ultrafast optical spectroscopy

Ultrafast optical spectroscopy offers an unmatched ability to differentiate the dynamics of spin, charge, and lattice, and the coupling between them in both time and spectral domains with femtosecond temporal resolution. CINT has a full range of time-integrated and time-resolved optical tools, covering terahertz through soft x-ray frequencies, to investigate the fundamental mechanisms of a wide range of nanostructured materials from physical to chemical to biological systems. A multitude of ultrafast experiments can be performed, including: optical pump-probe spectroscopy over the full frequency range, time-resolved photoemission spectroscopy and second harmonic generation, optical Kerr/Faraday spectroscopies, and ultrafast optical microscopy. All measurements may be done from 4 K to room temperature and under strong magnetic fields (up to 8 T). Examples of recent applications include studies of multiferroic magnetoelectric coupling dynamics, ultrafast carrier diffusion in individual semiconductor

nanowires, ultrafast photocurrents in topological semimetals, and ultrafast switching of metamaterials.

#### Advanced optical imaging and spectroscopy of single and multiple nanostructures

Optical characterization at the single nanostructure level is essential for revealing behaviors hidden in ensemble-level measurements and for providing spatially-correlated probes. CINT offers multiple scanning confocal laser microscopes for performing photoluminescence (PL) and PL excitation spectroscopies, as well as super-resolution, lifetime, and back-focal plane imaging operations. Direct wide area PL imaging is also available and provides simultaneous correlated imaging at multiple wavelengths. Instrumentation covers the spectral range from 350 nm to 1700 nm, and may be paired with a capability for controlling temperature (4-450 K) and gas and humidity of the sample environment. A time-correlated single photon counting capability also provides time-resolved PL and photon correlation/cross-correlation measurements using Si and InGaAs avalanche-photodiodes covering UV to near-IR wavelengths. Of particular note is a new EOS 410 superconducting nanowire single photon detector for ultrafast PL measurements in the near-IR. A cooled-cathode streak camera (temporal resolution  $\sim 2$  ps) and a fluorescence up-conversion system (temporal resolution  $\sim 150$  fsec) are also available for time-resolved PL measurements. This capability also includes instrumentation for performing magneto-PL and magneto-Raman spectroscopies to the single nanostructure level in magnetic fields up to 9 T.

#### Fully-tunable Raman spectroscopy and microscopy

CINT resonance Raman instrumentation provides a capability for materials characterization via vibrational fingerprinting, probing of electronic structure, electron-phonon coupling, and lattice response to external perturbation for a wide range of nanomaterials types. Employed as an imaging technique via integration of excitation sources to confocal imaging microscopes, we also provide spatial correlation with other spectroscopic imaging data to provide multi-modal analysis at bulk, thin-film, and single nanostructure levels. Our systems provide unique access to broadly tunable excitation sources, with continuous tunability from 345 nm to 1000 nm. Among the many materials studied with Raman at CINT, examples include carbon nanotubes, graphene and other 2D materials, SERS-active structures, bio and soft-material composites, quantum dots, nanowires, and multiferroic complex oxides.

#### 3D tracking microscope

Unique spatial filter geometry and active feedback in XYZ allows sub-diffraction limit measurement of the three-dimensional trajectories of fluorophore-labeled nanoscale objects moving at biologically relevant transport rates ( $\mu\text{m/s}$ ).

#### Bessel beam plane illumination microscopy

An alternative to conventional laser scanning confocal microscopy that enables rapid (100 frames per second) imaging in three-dimensions. Bessel beam plane illumination microscopy can be used for rapid 3D imaging of live mammalian cells and to study the 3D dynamics of select soft materials (e.g. diblock copolymer annealing, polymersome fusion).

### Super resolution fluorescence microscopy

This instrument provides optical images of fluorescently labeled samples at a spatial resolution of ~10-20 nm, approximately a factor of ten below the diffraction limit (~250 nm) and approaching that of electron microscopy.

### Scanning probe spatially correlated atomic force microscopy (AFM) and fluorescence imaging

A combination of optical spectroscopic and topographic information combines single-molecule fluorescence sensitivity with time-correlated single-photon counting (TCSPC) using pulsed laser excitation. Intensity or lifetime images of single emitters can be acquired and spatially registered with AFM images of the same area.

### Environmental scanning force microscopy

Scanning force microscope (Asylum) that can measure force between cantilever probe and surface providing information on adhesion and binding. Instrument features humidity control and an extended piezo with a Z-range of 40 mm.

### Mechanical/electrochemical/electrical scanning force microscopy

The Bruker Dimension Icon at CINT is comprehensively capable, with modules for Kelvin probe, tunneling, electrochemical, fluid sensing, complementary light-transmission experiments, electrical and magnetic modes, nanoindentation, and complete quantitative mechanical mapping in addition to the traditional contact and tapping modes. Many of these modes can be used simultaneously, and to complement many of the other CINT tools.

### Apertureless scanning near-field optical microscopy

Near-field microscopy overcomes the diffraction limit in optical imaging and spectroscopy, therefore representing an extremely important tool in CINT for the investigation of a host of integrated nanophotonic structures and devices. Combining atomic force microscopy (AFM) with optical imaging and spectroscopy, the apertureless scanning near-field optical microscope (a-SNOM) we acquired recently from neaSpec GmbH (Figure 4.6) allows a spatial resolution of 10-20 nm throughout the electromagnetic spectrum from far infrared (THz) to visible. Local optical properties can be examined via measuring the elastically scattered light resulting from the optical near-field interactions between the metal-coated or dielectric tip and the sample. It allows for subwavelength imaging across a broad portion of the EM spectrum, particularly in the mid-infrared wavelengths ranging from 4.7 to 15.3 microns, and the terahertz frequency range (0.3-3 THz). The a-SNOM also includes a pump-probe capability employing two femtosecond laser sources at 1560 nm and 780 nm. The capability is further enhanced by expertise provided by personnel in the Nanophotonics and Optical Nanomaterials thrust and the LUMOS team. This capability will allow us to investigate complex physical behaviors in systems including, but not limited to, strongly correlated materials, plasmonic response and wave propagation in 2D materials (e.g. graphene and quantum wells), nano-wires and carbon nanotubes, resonances in metamaterials, and other nanostructured functional photonic devices.



Figure 4.6 neaSPEC GmbH apertureless scanning near-field microscope.

### Holographic optical trapping

Non-contact manipulation of objects suspended in aqueous solutions using holographic optical trapping. The Arryx holographic optical trapping system allows the trapping and manipulation of small objects in 3 dimensions.

### Small- and wide-angle x-ray scattering

A commercial instrument (Bruker Nanostar) offers the possibility to rapidly evaluate structure of self-assembled soft materials on the Å to 100's of nm length scale. Sample temperature can be controlled from 5 °C – 85 °C.

## 4.4 Theory, simulation, and modelling

The unique properties of nanostructured and quantum materials cannot be fully exploited without a predictive understanding of the underlying phenomena. This requires a spectrum of theory/simulation techniques developed and optimized not only for the nanomaterial and quantum material component itself, but also for its interactions with surrounding components and materials. In addition to foundational capabilities like density functional theory (DFT) for electronic, optical, and vibrational properties of solids and nanostructures, some of the specialized techniques being used, under development, and planned include:

### Photoexcited dynamics

Among these tools, the Nonadiabatic EXcited-state Molecular Dynamics (NEXMD) framework developed by CINT scientists can efficiently and accurately describe photoinduced phenomena in extended molecular systems. It uses the fewest-switches surface hopping algorithm to treat quantum transitions among multiple adiabatic excited state potential energy surfaces. We achieve an accurate description of the multiple excited states by using the configuration interaction single formalism with a semi-empirical model Hamiltonian. The NEXMD methodology offers a computationally tractable route for simulating hundreds of atoms on ~10 ps time scales where multiple coupled excited states are involved. The software is approved at LANL for unlimited release for the broader scientific community.

### Molecular electronic structure database for machine learning (ML) algorithms

CINT is building a database of high-fidelity quantum-chemical computational results based on the extensive set of over 20 million molecular structures. The calculations were initially performed using a DFT approach and recently calculated in part with a very accurate Coupled Cluster (CCSD(T)) method. The inputs to the calculations are molecular geometries. The outputs contain molecular property information including molecular forces, molecular orbitals, orbital energies, electron densities, multipole moments, localized charges, bonding indices, energy decomposition, steric analysis, and resonance structures. The purpose of these data is to facilitate the construction of machine learning algorithms for energies and properties of molecular and solid state systems.

### Ultrafast quasiparticle dynamics

CINT is pioneering the theoretical modeling of ultrafast quasiparticle dynamics in strongly correlated electronic materials. In particular, our time-dependent Lanczos approach to electron-phonon and exciton-phonon coupling in solids keeps the full quantum nature of the problem. Our numerically exact approach has been shown to be many orders of magnitude more powerful

than other competing methods. It not only provides microscopic insight into the quasiparticle relaxation process but also guides the further development of phenomenological and effective modeling in complex materials. We will generalize this exact approach to treat the electronic correlation effects. We also plan to develop the time-dependent dynamical mean-field theory and density matrix renormalization group theoretical approaches to tackle correlation effects in high and low dimensional quantum material systems.

#### Electronic structure with correlations

CINT is also pioneering the theoretical modeling of local electronic structure with strong correlations. This capability has been applied to understand the bulk properties of correlated electronic materials by studying the electronic signatures around local impurities and defects. We have also developed quantum many-body approaches (Quantum Monte Carlo and Gutzwiller Variational Wave Function) into DFT-based first-principles method to tackle the problems of quantum impurities in an otherwise uncorrelated environment. The method will enable us to understand the material-specific localization and delocalization phenomena in quantum materials with strong electronic correlations. We have recently built a first-principles informed tight-binding modeling approach to model quantum materials at a larger scale and added a capability to analyze the topology of quantum materials. CINT has a track record in applying the GW method (where G = Green's function and W = screened Coulomb interaction) to study the quasiparticle excitation properties in solids, including actinides and the recently discovered perovskite solar-cell materials. We plan to develop a GW-BSE capability within a full-potential based electronic structure method, which enables us to treat more accurately the excitonic properties in semiconductors with strong spin-orbit coupling.

#### Model for soft materials and interfaces

CINT scientists have developed methods to model soft materials and the interface between soft and hard materials for systems such as polymer nanocomposites, biomolecule/materials and nanoparticle self-assembly using classical density functional theory, self-consistent field theory, molecular dynamics simulations, and Monte Carlo simulations. CINT scientist have contributed to LAMMPS, Sandia's highly parallel molecular dynamics code, particularly its capability to model nanoscale phenomena over multiple length scales through the introduction of efficient methods for modeling polymer nanoparticle composites, long range dipolar interactions, and atomistically inspired coarse graining of polymer. CINT staff have developed polymer self-consistent field theory codes to calculate self-assembled patterns and phase behavior of tethered polymeric systems. They also continue to develop Sandia's Tramonto code, a classical density functional theory code that describes equilibrium behavior of complex fluids in inhomogeneous environments, particularly at the nanoscale.

### 4.5 Leveraging capabilities

#### Laboratory for Ultrafast Materials and Optical Science (LUMOS)

The LUMOS facility<sup>5</sup> is equipped with ultrafast laser systems covering a broad spectral range that spans the far-infrared to the soft X-ray portion of the electromagnetic spectrum. These systems enable a multitude of ultrafast spectroscopic and imaging experiments, including optical-pump

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<sup>5</sup> <https://cint.lanl.gov/facilities/LUMOS/Capabilities.php>

THz-probe spectroscopy, high harmonic generation/extreme ultraviolet spectroscopies, and scanning probe imaging and spectroscopies. These capabilities enable us to understand and control the interaction of photons with the electronic, spin, and structural properties of materials on an ultrafast time scale for scientific and national security missions.

### [Ion Beam Materials Laboratory \(IBML\)](#)

The core of the IBML<sup>6</sup> consists of a 3 MV NEC tandem accelerator, a 200 kV Varian ion implanter, and a 200 kV Danfysik ion implanter together with several beam lines. The tandem and Varian have a joint target chamber for performing dual-beam ion implantation/irradiation experiments, which allows the simulation of neutron damage effects in reactors by concurrently performing self-ion irradiation (neutron displacement damage) and helium implantation effects (neutron induced nuclear reactions). The research capabilities include routine ion beam analysis techniques such as Rutherford backscattering spectrometry (RBS), nuclear reaction analysis (NRA), elastic recoil detection (ERD), particle-induced alpha- or gamma-ray emissions (PIXE or PIGE), and ion channeling, etc.; ion-enhanced synthesis and modification of materials through ion implantation; and ion irradiation damage effects in materials, including complex oxide ceramics, metals, semiconductors, and polymers. The coupled positron annihilation spectroscopy (PAS) to Tandem capability allows in-situ characterization of defect dynamics during ion irradiation. The coupled plasma to Tandem capability enables in-situ measurement of dynamic erosion rate of fusion plasma facing materials with ion beam analysis techniques or studying effects of irradiation induced defects on deuterium retention rate in plasma-facing materials. Ultra-low energy ions are able to do chemically doping in 2D materials.

### [Ion Beam Laboratory \(IBL\)](#)

The IBL<sup>7</sup> has a broad range of particle accelerators supporting fundamental and applied research. Compared to the broad beam capabilities at the IBML at the CINT-Gateway, the IBL at the CINT-Core provides users with focused ion beams, the beam spot size ranges from 10's nanometers to 10's micrometers, for targeted exposure of materials. The facility consists of four main accelerators that can produce ions from hydrogen to gold over a range of energies from electron volt (eV) to ~100 megaelectron volt (MeV) on target. The IBL has over 22 specialized end-stations enabling a wide range of experimental capabilities including focused ion implantation, in-situ electrical and optical testing, elevated and low temperature irradiations, in-situ SEM and TEM, and nanoimplantation.

### [Electrochemistry and Corrosion Laboratory](#)

The electrochemistry lab consists of a range of instrumentation for development or interfacing with many other CINT capabilities, including a broad range of Gamry and Bio-Logic potentiostats, specialty electrochemical power supplies, and electrochemical cells and related tools. The Gamry 600+ has a <250 ns rise time and current resolution of 20 atto-Amps, which fit sufficient for very small-scale localized corrosion investigations. The Bruker Dimension Icon atomic force microscopy (AFM) also has the electrochemical capability, with an electrochemical cell, temperature controller, and dedicated potentiostat. The developed electrochemical cell can be used for both the AFM and at beamlines to perform, for example, complementary investigations

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<sup>6</sup> <https://www.lanl.gov/science-innovation/science-facilities/ion-beam-materials-lab/index.php>

<sup>7</sup> [https://www.sandia.gov/m/research/facilities/technology\\_deployment\\_centers/ion\\_beam\\_lab](https://www.sandia.gov/m/research/facilities/technology_deployment_centers/ion_beam_lab)

leveraging the unique time and length scales of the AFM, the potentiostats, and beamline specific techniques such as phase contrast imaging. In addition, the electrochemical/corrosion cells fitted to loading platforms enable the study of stress corrosion cracking or the effects of cyclic loading.

### Microsystems Engineering and Science Applications Complex (MESA)

SNL has a significant role in advancing the state-of-the-art in microsystems research and development, and in introducing microsystems into the nuclear stockpile. Microsystems incorporate radiation-hardened microelectronics as well as other advanced components such as micromachines, optoelectronics, and photonic systems. The MESA Complex<sup>8</sup> (Figure 4.7) is designed to



Figure 4.7 MESA Complex.

integrate the numerous scientific disciplines necessary to produce functional, robust, integrated microsystems and represents the center of Sandia's investment in microsystems research, development, and prototyping activities. This suite of facilities encompasses approximately 400,000 square feet and includes cleanroom facilities, laboratories and offices. CINT currently leverages this NNSA resource by partnering with MESA staff in the design, development, and production of sophisticated CINT Discovery Platforms. We are planning to further leverage MESA by brining selected compound semiconductor synthesis capabilities into the CINT user program.

### 4.6 Future envisioned capabilities

Quantum research is at a critical threshold, with exciting opportunities for fundamental research as well as enormous potential for developing new quantum-based technologies. CINT is developing two new QIS capabilities: one focusing on the deterministic placement of quantum defects and the other developing quantum sensed nuclear magnetic resonance spectroscopy. Not only will this work bring many transformational advances in QIS research, the new capabilities will also enable the CINT user community to explore new frontiers in QIS, potentially opening the door to quantum information technologies that were previously considered impossible.

Solitary atomic defects capable of mimicking the quantum mechanical behaviors of individual atoms are considered the key materials for many revolutionary quantum information technologies. These defects can serve as fundamental building blocks for technologies ranging from ultra-sensitive magnetic field sensors to eavesdropping proof communication schemes and neuromorphic quantum computers. However, the realization of these technologies is hampered by the fact that researchers must primarily rely on serendipitously discovered, naturally formed defects. This means there is no control over the properties of the defects or their placement into devices. CINT aims to change that by developing two complementary strategies for 100% deterministic creation of defects that would also allow seamless integration of defects into nanoelectronic and photonic integrated circuits. The top-down strategy will be based on ion implantation technologies, expanded to demonstrate the deterministic creation of defects not only in bulk crystals (diamond and GaN) but also in low dimensional nanostructures (2D TMD and

<sup>8</sup> <https://www.sandia.gov/mesa/>

1D SWCNTs). The bottom-up strategy will involve the development of a new technology for ‘soft’ chemical implantation of quantum defects into 1D SWCNT and 2D VDWs.

CINT’s second QIS project will use a quantum qubit’s extreme sensitivity to magnetic fields to create a unique Quantum Sensed Nuclear Magnetic Resonance (QSNMR) Discovery Platform for magnetometry and nuclear magnetic resonance (NMR) spectroscopy at the smallest scale possible—a single spin—requiring orders of magnitude fewer spins than the most advanced techniques available today. The QSNMR Discovery Platform will be based on the fabrication of qubits using precise placement of nitrogen-vacancy centers in diamond substrates to serve as a quantum sensor for nanoscale NMR experiments. This will enable us to craft customized nanomagnetometer arrays that will allow CINT’s users, scientists, and research partners the ability to conduct NMR at unprecedented scale and resolution. This capability will enable the measurement of the magnetic properties inaccessible by any other means, in materials such as nanoparticles, topological materials, 2D atomic materials, polymers, single molecules, and biological materials.

In addition to these future QIS capabilities, we also envision developing capabilities in a number of other areas.

Femtosecond mid-IR microscopy is being planned to provide complementary capability to our other optical microscopies. Of particular interest will be opportunities for new studies in extended wavelength response and dynamics of emerging metamaterials and for probing of phonon coupling and control in opto-mechanical and heterostructured materials.

Expansion of single photon detection capabilities to the mid-IR is under construction using a combination of nonlinear up-conversion techniques. CINT will evaluate the procurement of single-photon superconducting nanowire detectors operating at longer wavelengths if they become available, which will complement our detectors operating in the near-IR.

CINT’s newest theoretical capability under development with users is a computational suite with DFT-informed tight-binding modeling of strongly correlated electron materials. It has the potential to include molecular dynamics with the tight-binding empirical parameters certified by DFT simulations. The method is targeted to address a plethora of emergent phenomena in transition-metal oxide nanocomposites.

In response to the rapid progress of hybrid structures involving nanoscale semiconductors and metals, we will expand our foundational single-band scattering matrix approach to develop a multiband theory of electronic and optical properties, and the charge and energy transport through the hybrid structures.

CINT has built and will soon make available to users a two-dimensional (2D) materials assembly system with user-friendly computerized controls in an inert atmosphere, providing users with translational and rotational degrees of freedom to fabricate architectures. Stacking and fabrication of 2D materials and their heterostructures in the 2D assembly system will be conducted by a control system in which real-time monitoring of 2D materials’ locations and XYZ-theta motorized movement are available. The inert atmosphere will be offered with a glove box for users to handle air-sensitive 2D materials, such as chromium iodides and tellurides. The assembly system will enable CINT users to perform various research projects encompassing

emerging nanoscience in twisted 2D layers and applications of stacked 2D materials for excitonic and high-mobility electronic devices.

To access longer time and length scales, CINT scientists are developing coarse grained models for treating soft materials on multi-length scales. These models along with enhancement to SNL's parallel molecular dynamics code LAMMPS to run on GPUs and the new high-performance architectures will provide unprecedented capabilities to model soft materials.

## 5 OPERATIONAL EXCELLENCE

The CINT management team is committed to operational excellence and draws on best-practice insights from both of the host labs. Prioritizing efficiency in business operations is one of the ways we can facilitate productivity for our user community. We will continue to explore new ways to position CINT as a user facility that meets the evolving scientific and operational demands of the nanoscience community.

The CINT user program is designed to provide the international scientific community access to the world-class capabilities and scientific expertise available at the CINT Core and Gateway facilities. Access is typically requested through a biannual, peer-reviewed proposal process. Approved user projects are valid for 18 months, during which time users can access the facilities as needed. In addition, Rapid Access proposals can be submitted in the periods between the biannual calls. This provides a mechanism for a prospective user to have limited access to CINT capabilities in advance of the next call for user proposals for projects that are time-critical, small in scope, and high-impact. We regularly assess the effectiveness of our user program to ensure the proposal process is streamlined.

One area of planned improvements is our user program database. With over 800 users accessing the CINT user program in FY19, it is critical that we have a state-of-the-art database for managing proposal submissions, capability allocations, onsite visits, and productivity metrics. Our discussions with the LANL and SNL User Facilities Working Group, the other NSRCs, and other members of the Society for Science at User Research Facilities<sup>9</sup> have been a valuable forum for benchmarking best practices in user program management. We are also preparing a series of updates to the NSRC Portal<sup>10</sup>, a community website featuring a capabilities database and science highlights from the NSRCs. The Portal is a valuable outreach tool for the NSRCs as it enables users to explore capabilities and expertise across all of the centers. We envision the next-generation NSRC Portal to not only feature this comprehensive search function, but also be a hub for user interactions across the centers.

Safety is an integral part of CINT's operation. By utilizing continuous feedback through our Integrated Safety Management approach, we are constantly evaluating and improving our ES&H operations as well as the training and qualifications of our staff and users. The ES&H professionals from the Core and Gateway facilities have both performed site visits to familiarize themselves with the different host labs' approaches and have shared experiences on how to improve and unify our procedures. We have also implemented peer walkthroughs to learn from the different approaches the two laboratories take in their safety programs. These walkthroughs are intended as learning opportunities rather than oversight and staff is encouraged to share experiences openly without written documentation. In this way we aim to create a safety culture where staff and users feel empowered to be advocates for a safe and secure workplace.

We have the opportunity to lead the complex in improving cross-laboratory processes, in particular with our activity in creating a safety-training program that is acknowledged at both LANL and SNL. Through CINT's ES&H team, LANL and SNL have jointly standardized the NANO101

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<sup>9</sup> <http://www.ssurf.org/>

<sup>10</sup> <https://nsrcportal.sandia.gov/>

and RAD210 trainings. Each lab has also created granting mechanisms to recognize and document these equivalencies. Our ES&H professionals continue to work with the host laboratories to approve reciprocity for additional required trainings that are required for our users accessing both sites.

## 6 ENGAGING THE SCIENTIFIC COMMUNITY

Realizing our vision of nanoscience integration requires partnerships between many members of the scientific community. CINT is part of a network that spans our staff, users, host labs, the NSRCs and BES, as well as the larger science community. These relationships, and the roles we play in each relationship, are vital to our success as a user facility. As such, we are committed to supporting CINT's scientific staff to be leaders in their fields and to growing an engaged user community.

Scientific leadership is one of the ways we promote our differentiating capabilities and attract active users. Our most recent user satisfaction survey revealed that more than 50% of respondents heard about CINT through our staff or a CINT publication, highlighting the important ambassadorial role our scientists play. Our staff and users take on leadership roles by organizing conferences, workshops, and symposia and by serving the community in the capacity of journal editors, reviewers, and in service to professional organizations. Invited talks at conferences and universities are another example: Between 2016 and 2018 our staff and users gave over 400 invited talks.

To ensure that we can support our existing staff in these endeavors, as well as attract high-achieving new staff, we are committed to improving professional development opportunities and early-career mentoring. Strategic hires of scientists, technologists, and postdoctoral researchers will be based on BES initiatives, the needs of the nanoscience community, and addressing our integration challenges. In the context of hiring and human resources, we also affirm our commitment to diversity, equity, and inclusion. These are defining elements in the workplace cultures at LANL and SNL; these qualities foster multiple perspectives and encourage innovation, both of which are crucial for achieving our science vision.

In a similar vein, one of our main goals is to expand our user community by attracting research leaders from diverse institutions, early career scientists, and innovators in nanotechnology companies. This requires targeted outreach activities to increase awareness of our capabilities and expertise. Our targeted efforts include continued presence in high-profile journals and organize symposia at national meetings and workshops on nanomaterials integration challenges. We will continue to complement these activities by partnering with the other NSRCs to increase our presence at national meetings. For the 2019 MRS Fall Meeting & Exhibit, which attracted over 6000 materials science researchers, we worked with the MRS exhibits manager to highlight user facilities throughout the meeting. This included an exhibit row dedicated to user facilities. The NSRC booth (Figure 6.1) was one of 16 booths in the row representing over 100 user facilities. In addition to the exhibit, user facilities were featured in panels, workshops, and Hub Stage presentations. Participating in national meetings in this way allows us to maximize our impact and complements the technical



Figure 6.1 The NSRC booth at the 2019 MRS Fall Meeting & Exhibit.

programs led by CINT staff and users. We will continue to explore new ways to capitalize on the NSRC presence at conferences and workshops.

CINT has maintained strong ties to industry researchers, both from local small business and global companies; we have consistently had 6-8% of our users from the private sector. To further encourage industrial participation, we plan to identify and reach this community by more strongly leveraging the SAC and UEC, better engaging our host laboratories' tech transfer offices to see how industry challenges can be aligned with CINT capabilities, working with state and local economic development organizations, and more actively showcasing our capabilities at industry-targeted conferences and workshops.

We draw on the pool of dedicated scientists who make up CINT's User Executive Committee (UEC) and Scientific Advisory Committee (SAC) to not only provide representation for all our users but also to ensure that CINT is well positioned to respond to emerging trends in nanomaterials integration. The UEC provides a voice for our users and an organized framework for communicating with CINT management. The UEC helps organize the CINT Annual Meeting, which is a venue for demonstrating to current and potential CINT users the range of CINT capabilities enabling cutting-edge research in nanoscience integration. The meeting also highlights user research performed at CINT and is a forum for discussing strategic plans with the community through our focused symposia. The SAC is composed of external scientists who provide guidance in areas such as: evaluating scientific programs; providing advice on future science directions and infrastructure needs, and; ensuring that CINT maintains a highly effective user program. We will continue to work closely with the SAC and UEC to ensure open communication channels between users, user representatives, and the CINT leadership team.

We value the in-person interactions we have with our staff, SAC, and UEC, and it is also important to maintain open and active communication channels with current and potential users. To this end, we maintain a regularly updated website<sup>11</sup> and highlight current news on both the website and social media platforms. We have greatly increased our social media reach by developing relationships with the LANL and SNL media teams; CINT content that is highlighted on their accounts has an audience of over one hundred thousand social media users. In terms of our promotional materials, we have diversified our offerings with the addition of a CINT comic and continue to work with our host labs' graphic design and media teams to explore new ways to promote CINT to a wide audience. We also encourage all of our users to complete the Annual User Satisfaction Survey. We have seen increases in the number of respondents each year and will continue to analyze user input to make technical and operational improvements.

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<sup>11</sup> <https://cint.lanl.gov/>

## 7 SUMMARY AND IMPLEMENTATION

CINT2026 presents a vision for potentially revolutionary new technologies that would be enabled by integrating nanostructured materials to exploit their size-dependent properties or emergent collective properties. Realization of this opportunity requires a sustained effort by researchers to tackle more complex nanosystems; however, the optimal methods to do so may not necessarily exist today or may not be available to researchers with extraordinary ideas and insights.

CINT's differentiating focus on nanomaterials integration and established operation as a DOE Office of Science user facility with a vibrant, growing user community, position us to be a leader at this new frontier in nanoscience. Our six strategic objectives, listed in Section 2, drive us towards the goal of being the national resource for research expertise and unique capabilities to synthesize, fabricate, characterize, and understand nanostructured materials in increasingly complex integrated environments.

This strategy involves harnessing the intellectual leadership and scientific expertise of our staff and users (objectives 1 and 6) by adding CINT scientists and attracting international experts as CINT users. A second strategy complements the people in our community with the capabilities that we invent or optimize for integration research (objectives 2, 3 and 4), thereby ensuring that CINT is unsurpassed as an institution for our chosen areas of nanoscience. In an increasingly competitive world, CINT must also continuously focus its efforts and resources on the potentially most impactful research opportunities (objective 5). This will be done through organized CINT workshops, the CINT Annual Meeting, joint-NSRC workshops, focused symposia at major national scientific conferences, and consultation with the CINT Scientific Advisory Committee and the CINT Users Executive Committee.

When the National Nanotechnology Initiative was launched over 15 years ago, the nation was at the dawn of the next technological revolution. Just as integration transformed the transistor into the integrated circuit, now the scientific community is poised to reveal even greater functionality by learning how to build a world of multifunctional materials and systems that can begin to rival the exquisite examples we see every day in nature. Our vision for CINT in the year 2026 ensures that we will be leaders in transforming our understanding of nature and strengthening the connection between advances in fundamental science and technology innovation.