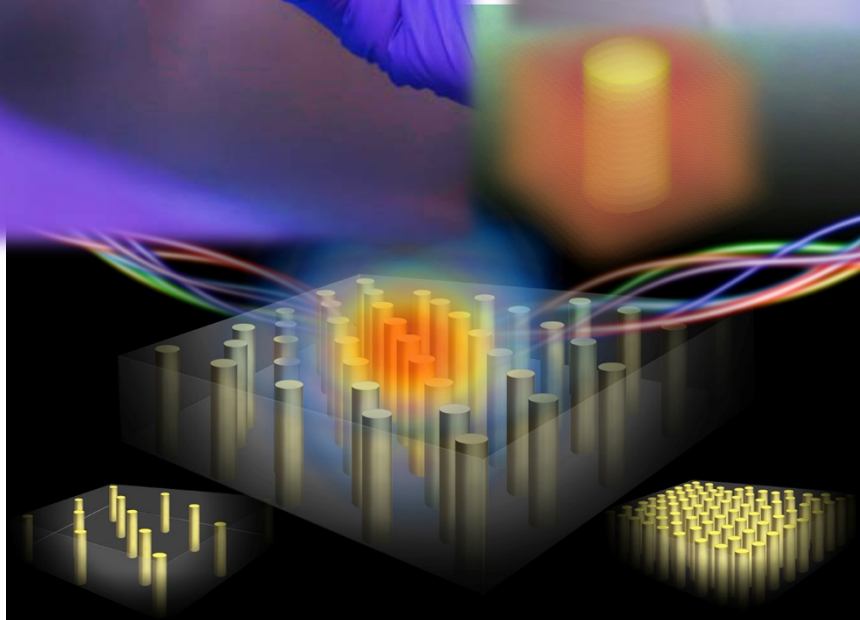
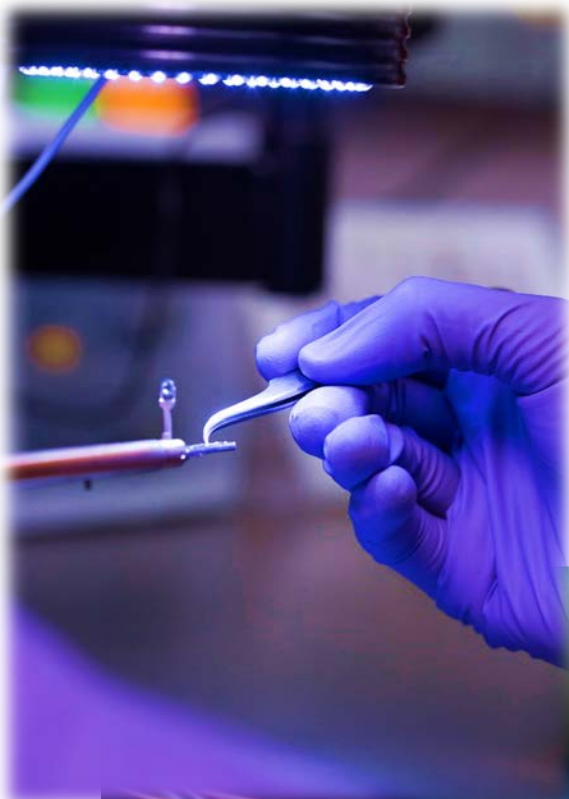




# CINT 2022



## The 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, operating 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 to Los Alamos Facility, CINT provides access to *scientific expertise* and *advanced capabilities* for researchers to synthesize, fabricate, characterize, understand and integrate 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 Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000. Los Alamos National Laboratory, an affirmative action equal opportunity employer, is operated by Los Alamos National Security, LLC, for the National Nuclear Security Administration of the U.S. Department of Energy under contract DE-AC52-06NA25396.

## Executive Summary

The Center for Integrated Nanotechnologies (CINT) plays a leadership role in the area of integration science through its function as a Department of Energy/Office of Science Nanoscale Science Research Center (NSRC) national user facility. 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 that which is achievable by individual researchers or any single institution.

Deriving the ultimate benefit from nanoscience will require the assembly of diverse nanoscale materials across multiple length scales to design and achieve new properties and functionality; in other words, nanomaterials integration. Integration has played a pivotal and revolutionary role in the development of nearly all science and technology and is the overarching theme of CINT. Our strategic plan is based upon the extraordinary scientific opportunities involving nanomaterials integration in three challenges inspired by CINT users and our staff. Solving challenges such as these would unlock great possibilities for innovative technologies and have widely recognized impact in nationally important areas such as energy, environment, human 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 (LANL) and Sandia National Laboratories (SNL), 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 to benefit hundreds of researchers annually. We have world-leading *scientific expertise* in four thrust areas, as described in section 1, 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 that can be used alone or as sophisticated “experiments within an experiment”; 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-high resolution spectroscopic techniques of nanomaterial dynamics; *in situ* microscopies that provide real-time, spatially-resolved structure/property information for increasingly complex materials systems; advanced simulation techniques for integrated nanomaterials; and multi-scale theory for interfaces and dynamics.

CINT's transformational and supporting foundational tools and 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 will target research leaders in diverse institutions, early career scientists, and innovators in nanotechnology industries to create an expanded national user community of the future.

Finally, the CINT2022 vision requires not only the prudent allocation of precious federal resources to support operations but also mutually beneficial new partnerships to bring state-of-the-art expertise and capabilities into the user facility of the future. We embark on this exciting journey ever mindful that the most significant metric of success is the quality of the science and technology produced by the CINT community.

## Contents

<u>CINT MISSION .....</u>	<u>I</u>
<u>EXECUTIVE SUMMARY .....</u>	<u>II</u>
<u>LIST OF FIGURES.....</u>	<u>IV</u>
<u>1.0 INTRODUCTION, STRATEGIC OBJECTIVES AND ORGANIZATIONAL ALIGNMENT.....</u>	<u>1</u>
2.1 INNOVATIVE NANOFABRICATION, INTEGRATION, AND “UP-SCALING” METHODS TO INCORPORATE QUANTUM-SIZE NANOSTRUCTURES INTO ARBITRARY 2D AND 3D ARCHITECTURES .....	4
2.2 HYBRID MATERIAL INTERACTIONS FOR GENERATION AND MANIPULATION OF LIGHT .....	8
<u>3.0 SCIENTIFIC EXPERTISE TO REALIZE THE VISION .....</u>	<u>15</u>
4.1 DISCOVERY PLATFORMS: A CINT SIGNATURE INITIATIVE.....	17
4.2 SYNTHESIS / FABRICATION .....	19
4.3 CHARACTERIZATION.....	22
4.4 THEORY, SIMULATION AND MODELING.....	25
4.5 LEVERAGING CAPABILITIES .....	26
4.6 FUTURE ENVISIONED CAPABILITIES.....	27
<u>5.0 ENGAGING THE NATIONAL/INTERNATIONAL SCIENTIFIC COMMUNITY .....</u>	<u>29</u>
<u>6.0 OPERATIONAL EXCELLENCE.....</u>	<u>29</u>
<u>7.0 IMPLEMENTATION .....</u>	<u>31</u>

## List of Figures

Figure 1. CINT leverages its diverse scientific community to fully integrate theory and experiment.....	4
Figure 2. a) Thin films, (b) a homogeneous nanowire array, and (c) an hypothetical.....	5
Figure 3. Nanowire Arrays.....	6
Figure 4. Example non-classical photon emitters arising from engineered materials interactions. ....	8
Figure 5. Integrated meta-molecule based metamaterials. ....	9
Figure 6. (Left panel) 3D trajectory of a single allergy antibody (IgE) labeled with a single gQD on a plasma membrane. (Right panel) White light image of the cell during the trajectory map. Ref. Keller et al. 2014 Adv. Func. Matter. ....	12
Figure 7. Water-induced swelling and contraction of a tetragonally perforated lamellar structured polymer serves to regulate macroscopic optical response of spatially organized plasmonic NPs (Au) and emitters (gQDs).....	12
Figure 8. Addressing integration through collaborative capabilities and expertise in fabrication, synthesis, characterization and theory.....	17
Figure 9. Electrochemical Discovery Platform.....	18
Figure 10. Microfluidics Discovery Platform.....	18
Figure 11. Atomic Precision Lithography.....	20
Figure 12. Dip Pen Nanolithography using a quantum dot ink.....	21
Figure 13. R&D 100 award winning 3D Tracking Microscope.....	24
Figure 14. neaSPEC GmbH apertureless scanning near field microscope.....	24
Figure 15. A polaron quasiparticle (an electron dressed by fully quantum phonons) in a nanowire is excited by an ultrafast optical pulse.....	25

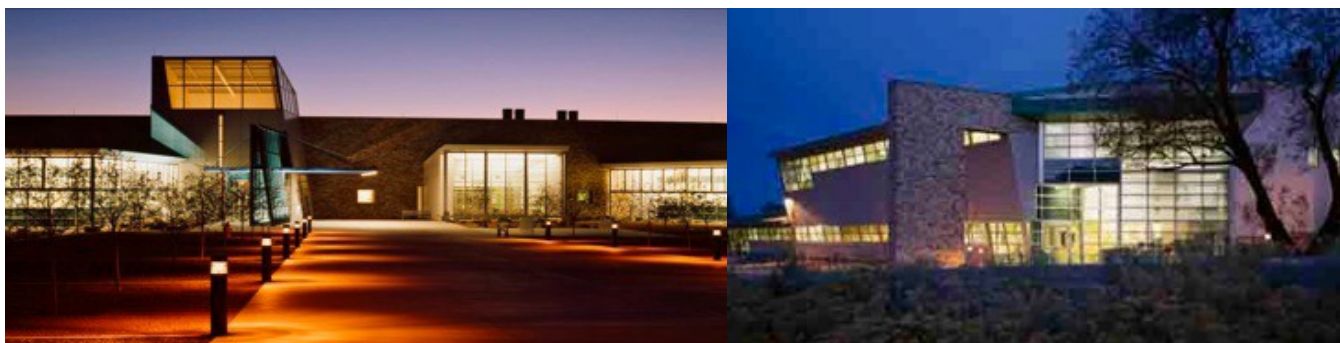


## 1.0 Introduction, Strategic Objectives and Organizational Alignment

The Center for Integrated Nanotechnologies (CINT) is a Department of Energy/Office of Science Nanoscale Science Research Center (NSRC) operating as national user facility. As a vibrant partnership between Los Alamos and Sandia national laboratories, CINT leverages the unmatched scientific and engineering expertise as well as special capabilities of our host DOE Laboratories to create 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. 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.

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Thus our overarching goal:



To be the national resource for *research expertise* and *advanced capabilities* to synthesize, fabricate, characterize, understand and **integrate** nanostructured materials in order to achieve unprecedented materials functionality and innovated systems and inspire revolutionary nanotechnologies.

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

1. CINT will be the nationally recognized leader in nanomaterials integration.
2. CINT will develop unique experimental and theoretical capabilities to synthesize, fabricate, characterize, and understand nanoscale materials in *increasing complex, integrated environments*.
3. CINT will develop next generation Discovery Platforms™ inspired by nanoscience integration challenges and will seek to proliferate the use of Discovery Platforms™ at user facilities and other institutions nationwide.
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 capability 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. Department of Energy’s Strategic Plan 2014-2018. Specifically:

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DOE Strategic Objective 3 – 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.

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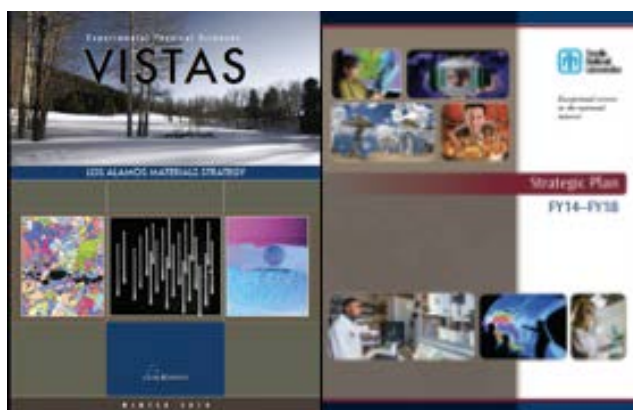


In order to accomplish DOE Strategic Objective 3, the Department of Energy has three strategies, each of which is supported by CINT:

1. Conduct discovery-focused research to increase our understanding of matter, materials and their properties through partnerships with universities, national laboratories, and industry.
2. Provide the nation’s researchers with world-class scientific user facilities that enable mission-focused research and advance scientific discovery.
3. Use the national laboratory system and leverage partnerships with universities and industry to conduct mission-focused research.

CINT is already implementing the three DOE strategies above. We have world-leading *scientific expertise* in and *specialized capabilities* to synthesize, fabricate, characterize and understand nanomaterials in increasingly complex integrated environments. Our expertise is organized in four scientific thrust areas:

- *Nanophotonics & Optical Nanomaterials* - Synthesis, excitation and energy transformations of optically active nanomaterials and collective or emergent electromagnetic phenomena (plasmonics, metamaterials, photonic lattices).
- *Nanoscale Electronics & Mechanics* - Control of electronic transport and wave functions, and mechanical coupling and properties using nanomaterials and integrated structures.
- *Soft, Biological & Composite Nanomaterials* - Solution-based materials synthesis and assembly of soft, composite and artificial bio-mimetic nanosystems.
- *Theory & Simulation of Nanoscale Phenomena* - Assembly, interfacial interactions, and emergent properties of nanoscale systems, including their electronic, magnetic, and optical properties.



In addition, we leverage the strengths of our host DOE laboratories through the alignment of the CINT 2022 strategic plan with the LANL and SNL missions, capabilities and strategic investments. For example, through the exploration of materials and integration, we support the LANL “Materials for 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 relevant to national security missions. Similarly, the SNL Strategic Plan includes explicit

objectives in support of the nanoscience capability, materials science research foundation, and multiple crosscutting research challenges involving the creation, understanding and integration of advanced nanomaterials in technologies for national security applications.

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 250 host facility researchers annually at the facilities, 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 LDRD), 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 three 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; 2016 - Transceiver for Quantum Keys and Encryption (R.M. Camacho, J. Urayama\*, P. Davids\*, et. al.), 2016 - Stress-induced Fabrication of Functionally Designed Nanomaterials (Hongyou Fan\*, Willie Luk, Igal Brener, et. al.), and 2014 - Nano-Enabled, Microfluidic Detection of *Bacillus anthracis* (Harper\*, Bachand). BaDx (Bacillus anthracis Diagnostics) is recognized as a 2015 TechConnect Innovation Awardee at the TechConnect National Innovation Summit. It has been selected as the Best Tech of the Year by Popular Science (December, 2015). This technology has been dubbed as the “smallest, safest Anthrax detector.” It won the Grand Winner Award under category of Security. These achievements illustrate the ever-growing potential for CINT to amplify the exceptional nanoscience strengths of our host DOE laboratories. (\* CINT users)

Finally, we note the special role that the DOE Office of Science user facilities have in supporting key objectives in 2014 National Nanotechnology Initiative Strategic Plan:

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3.3. Provide, facilitate the sharing of, and sustain the physical R&D infrastructure, notably user facilities and cooperative research centers.

3.3.1. Establish regular mechanisms to determine the current and future infrastructure needs of users and stakeholders of these facilities and centers.

3.3.2. Develop, operate, and sustain advanced tools, infrastructure, and user facilities (including ongoing investment, staffing, and upgrades).

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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, human health and security. We describe three representative challenges in section 2, followed by the requisite expertise and capability development in sections 3 and 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 2020, the six CINT strategic objectives that are the pathway to achieving the vision, and dedication to continued excellence as a DOE Office of Science scientific user facility ensure that CINT will transform our understanding of nature and strengthen the connection between advances in fundamental science and technology innovation.



## 2.0 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 amplify their properties (up-scaling) or lead to new ensemble behaviors (emergent phenomena). 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 (or computer “chip”) did. The development of the “chip” required the capability to integrate a large number of resistors, capacitors, diodes, and transistors on a single platform. Once the “chip” 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 CINT2022 Strategic Plan builds upon this foundation by illustrating three 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 three challenges is a fully integrated feedback loop of synthesis, fabrication, characterization, and modeling, shown in Figure 1 (also described in more detail on the [NSRC Portal](#)) 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 1. CINT leverages its diverse scientific community to fully integrate theory and experiment.



### 2.1 Innovative nanofabrication, integration, and “up-scaling” methods to incorporate quantum-size nanostructures into arbitrary 2D and 3D architectures

**Scientific opportunity:** Semiconductor nanowires and nanotubes are important building blocks for next-generation energy-harvesting and energy-storage systems, optoelectronics (from single-photon sources to low-threshold lasers), photodetectors, and even sensors for chemical or biological agents. Significant progress has been made in recent years in the precision bottom-up synthesis of single-crystalline, controllably doped, and heterostructured (both radially and axially) nanowires, as well as in the selective preparation of nanotubes with specified structure

and chirality. In addition, substantial improvements have been made in top-down techniques in 2D heterostructured thin films to fabricate, grow, and understand nanomaterials with desired electronic and optoelectronic properties that could lead to future innovation for energy-efficient light capture (e.g., solar energy, light/radiation detection) and light emission (e.g., solid state lighting, quantum communication). CINT is already at the frontier of these research areas, and is perfectly poised to address the challenges in:

- Innovation in nanofabrication and integration of the growth of arbitrary, 3D, multifunctional, quantum-size structures.
- Integration and “up-scaling” methodologies for transforming 1D nanowires and nanotubes into 2D and 3D architectures and ensemble systems designed for advanced functionality.

**Positional science and capabilities:** CINT has state-of-the-art capabilities in molecular beam epitaxy (MBE) growth of high-mobility III-V planar heterostructures (see section 4.2). CINT’s MBE is in demand worldwide to grow high-purity, ultra-high mobility As-based III-V compound semiconductor structures with atomic monolayer precision for fundamental studies of 1-D and 2-D nanomaterials. CINT has a 9,000ft<sup>2</sup> cleanroom facility with processing tools that can reach a resolution of tens of nanometers. CINT specializes in the synthesis of semiconductor nanowires by solution-phase and chemical vapor deposition (CVD) approaches to produce single crystal nanowires, radially and axially heterostructured nanowires, and complex architectures consisting of Si/Ge, III-V and other compound semiconductor materials. Hybrid 3D nanostructures are being fabricated by growing Si and Ge nanowires and their heterostructures on 2D graphene and transition metal dichalcogenides. The capability to modulate the catalyst composition in-situ in the Si-Ge CVD system enables precise control of nanomaterial characteristics. The CINT nanomanipulator is a custom two-probe device inside of a scanning electron microscope, and is used for in-situ quantitative nanostructure electrical characterization as well as the fabrication of single nanowire devices for ex-situ electrical, thermal, and optical property measurements. CINT’s Discovery Platform for in-situ TEM measurements of electrochemical processes in single nanowires has enabled many discoveries in this area. For example, silicon nanowire structures are attractive for use in next-generation energy-storage systems for their high energy density as negative electrodes for Li-ion batteries. The high surface area of the nanowire structures allows for the accommodation of the 300% volume expansion during charging. In scaling up these materials, CINT researchers are investigating the degradation mechanisms of silicon nanowire arrays in comparison to degradation within individual amorphous and single crystalline structures, and are developing design strategies to mitigate the capacity loss that occurs when active materials are disconnected from the current collector. CINT has demonstrated the application of ultrafast wide field optical microscopy for multi-scale characterization, and has routinely used ultrafast pump-probe optical spectroscopies to study the carrier behavior in single nanowires.

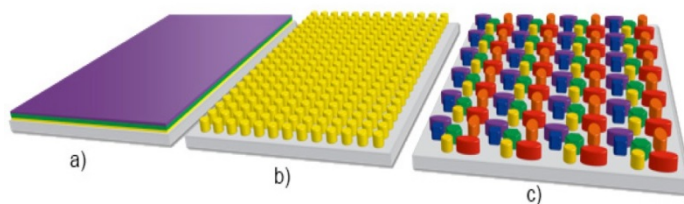


Figure 2. a) Thin films, (b) a homogeneous nanowire array, and (c) an hypothetical.

**Toward the future:** 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 utility to many technologies including consumer

electronics, energy, and biomedicine (Figure 2). *However, this will require substantial innovation in the nanofabrication, growth, and integration of quantum-size structures.*

Combinations of bottom-up and top-down growth and nanofabrication techniques have been used successfully to create “vertical” nanowires (Figure 3) in a few material systems such as group IV and certain III-V semiconductors. More complex 3D structures that contain a variety of semiconductors with sections of varying dimensional confinement (1D, 2D & 3D) are required for new applications such as low threshold lasers, quantum information and computation, new transistor architectures that go beyond the semiconductor roadmap, and novel optomechanical systems. To realize this vision, hybrid multi-sequential combinations of bottom-up and top-down synthesis techniques must be developed in a variety of combined semiconductor families, possibly oxides or diamond. One hypothetical example of an application where heterogeneous integration of dissimilar structures and materials could be used is single-photon sources for quantum information processing. An ideal single-photon-on-demand source could be made from an electrically injected quantum dot. One option for accomplishing this is to fabricate a nanodevice made from different direct bandgap III-V semiconductors emitting at different wavelengths. The semiconductors would have to be placed in specific locations in a 3D arrangement for subsequent coupling to passive waveguides made from group IV semiconductors for further multiplexing and routing. Developing architectures such as this will require the integration of different dimensional structures into a larger functional microstructure.

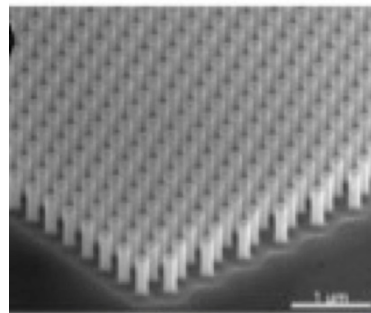


Figure 3. Nanowire Arrays.

CINT is looking to expand its growth capabilities into other semiconductor families such as large band gap semiconductors (i.e., III-Nitrides, diamond) and low band gap materials (e.g., InGaAs/InP, antimonides), and expand the MBE effort into high-mobility group IV materials. The integration of different semiconductor materials laterally and vertically will necessitate the development of new hybrid growth techniques with *in situ* sample handling and characterization in vacuum, all combined with new nanofabrication techniques. In addition to the expansion and combination of epitaxial capabilities, new nanofabrication techniques must be developed to create arbitrary top down nanostructures that go beyond “vertical etching.” CINT researchers envision the capability to create quantum wires that lie horizontally in several planes, and with arbitrary control of their diameters and connectivity to other top-down or bottom-up defined sections.

CINT has exceptional capabilities in synthesis and characterization of individual nanowires. Probes have been developed that make it possible to determine electrical, optical, and thermal properties at the level of a single nanowire or nanotube, and even physically manipulate single wires. *However, one of the biggest challenges in nanowire science is how to integrate, up-scale, and organize structures and architectures into ensemble systems that are functionally relevant.*

CINT is well positioned to answer this challenge by leveraging established capabilities in nanowire and nanotube synthesis and chemical modification, nano-manipulation for direct measurement of a nanostructure’s properties, and fabrication of single nanowire devices in order to address the fundamental issues of integration:

- manage the interface effects between individual nanostructures in interconnected networks and composite mesoscale structures;

- bridge the gap between emergent nanoscale functionality and macroscale performance;
- develop architectures that harness the exceptional axial diffusion behaviors of 1D systems;
- and understand the fundamental processes impacting ion and electron transport behaviors at the junctions of nanostructures.

CINT will expand the Discovery Platforms for mechanical / liquid environments and electrical in-situ TEM measurements of the environmental impacts on transport electrochemical and mechanical processes in single nanowires and their interfaces. Through understanding the core degradation mechanisms that plague nanowire arrays, CINT researchers are developing design rules to implement these structures into bulk electrodes for integration into Li-ion energy-storage systems. The electrochemical TEM Discovery Platform will be redeveloped for enhanced environmental control for novel operando testing to expand the “lab-in-a-gap” capabilities. CINT will develop unprecedented capabilities in multi-scale characterization, including integrating wide field observation with nano- and mesoscale resolution in up-scaled three-dimensional architectures.

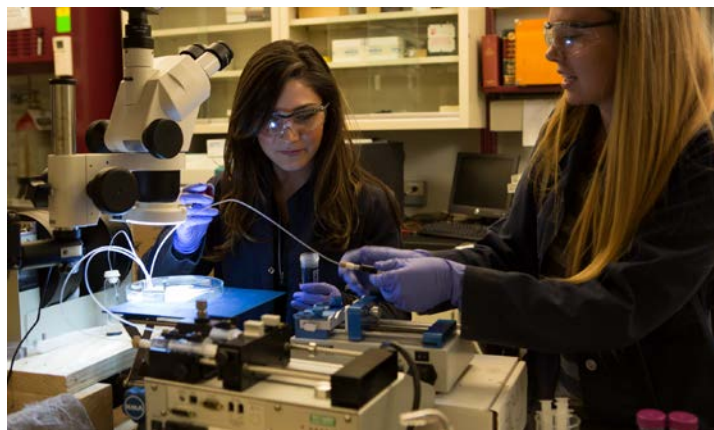
*Meeting the modeling challenge:* CINT’s ultimate goal in this area is to develop a fundamental understanding of the interfacial interactions between nanoscale components with 2D and 3D confinement (i.e., nanowires and quantum dots) and the host materials, and how these interfacial interactions affect the overall functionality of the hybrid structures, thus leading to optimization and control by design. CINT will pursue the following directions:

- Develop a predictive capability for the relationship between interfacial structure (microstructure and vacancies/defects) and carrier transport across interfaces. This goal will be accomplished through first-principles calculations of structural instability at interfaces and the functionality, including local electronic structure and transport, across the interfaces.
- Develop a new theoretical framework to address transport properties across entire hybrid structures at the ensemble scale. This framework will be based on the first-principles calculations for both individual nanowires/QDs and the interfaces and will help establish the relation between the structure functionality and electronic, optical, and transport properties at the mesoscale.



## 2.2 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 novel photon generation and manipulation. 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:



- Materials and structures that control and modify electromagnetic energy (plasmonics, metamaterials); and
- 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, their characterization with powerful spectroscopic tools, paired with an ability to control compositions and assembly routes to define interaction geometries across multiple length scales and degrees of complexity. We are international leaders in developing exceptional photonic materials with switchable and highly tunable photon emission properties. Examples include proprietary non-blinking quantum dots and novel microfluidic control of synthesis for axially heterostructured nanowires. Our doped carbon nanotubes provide new multifunctionality and boosted quantum yields and highlight CINT ability to isolate specific tube structures and control their surface chemistries. Together, our emitters provide multi-photon to single photon behaviors across classical to quantum regimes (see Figure 4). Pioneering efforts in nanomaterials assembly include soft templating approaches for creation of hybrid functional systems with hierarchical structures that are reconfigurable and responsive. Innovative dip pen nanolithography is providing unprecedented control over placement of emitters on photonic, plasmonic, and metamaterials structures, which provide significant opportunity for manipulation of light. CINT has been a world leader in the area of metamaterials (THz to near-infrared) for nearly a decade. We now lead the world in all-dielectric metamaterials as well. CINT's position in this field is enabled by access to nanofabrication and epitaxial growth facilities.

Advances in these materials are driven by world-class spectroscopic characterization, including ultrafast tools providing fs resolution across THz to soft X-ray energies unavailable anywhere else. 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 abilities for

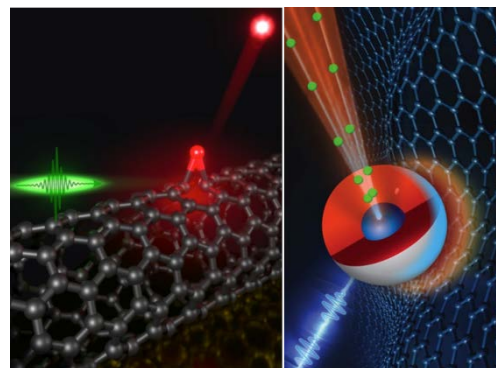


Figure 4. Example non-classical photon emitters arising from engineered materials interactions.



optical characterization of nanomaterials are unmatched. Our first-principles DFT simulation capability encompasses nearly all flavors of electronic structure codes for understanding electronic, optical, and vibrational properties of complex materials. In particular, 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 simulation for metamaterials modeling and design.

**Toward the future:** Drawing on these strengths, CINT will approach 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 the 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.
- Synthesizing and/or integrating multi-component systems with the appropriate interaction geometries to create a desired functionality, such as harnessing metamaterial interactions with dopant states of emitters for directional emission or enhanced coupling to photonic waveguides.
- Generating desired optical responses in emergent electronic structures by manipulating interactions across multiple length-scales within interfacial environments. Examples include use of soft responsive systems to modulate coupling between embedded optical emitters for on-demand behaviors.

CINT will employ a variety of materials processing techniques, including direct synthesis, self-assembly, nanofabrication and directed placement, to further our understanding of these issues. CINT will also move beyond traditional synthetic processing approaches by tapping soft-materials assembly methods with the potential 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

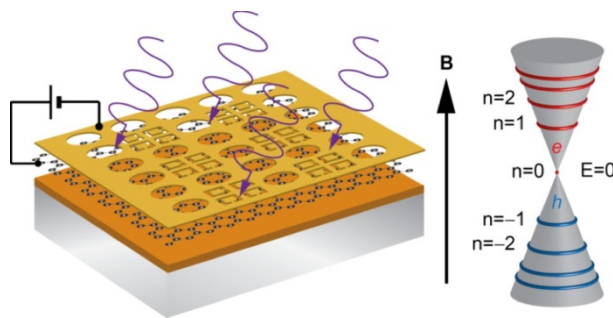


Figure 5. Integrated meta-molecule based metamaterials.

nonlinearities in metamaterials coupled to different materials. 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. To realize this extraordinary multifunctionality, CINT will utilize our integrated efforts in synthesis, characterization, and modeling to address the following challenges:

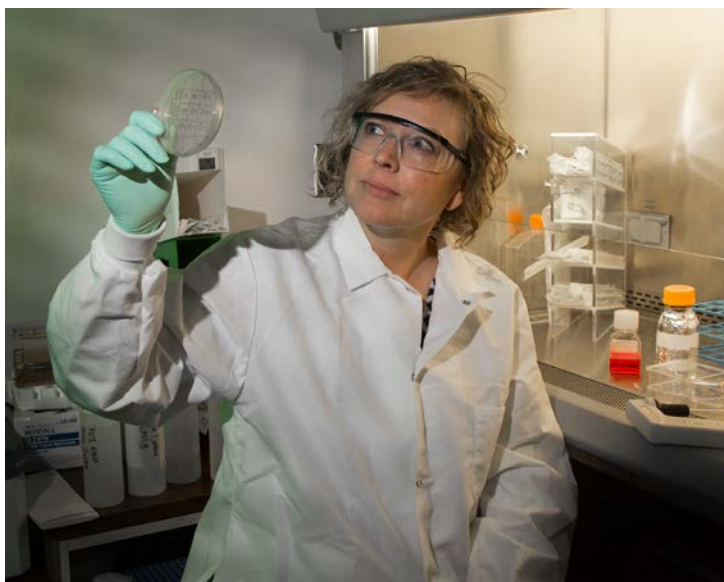
- Devising non-traditional plasmonic systems (e.g., graphene hybrids or emerging epitaxial oxides) that cannot be accessed with more traditional noble metal approaches.
- Designing multifunctional metamaterial behaviors through metamolecule concepts (shown in Figure 5).
- Generating hybrid interactions coupled to metamaterials architectures to provide active/dynamic control and tuning of enhanced metamaterial response and their optical nonlinearities.

*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. 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, Dirac metals) 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. However, the full range of effort in this area will require CINT to expand its materials generation capability to include new techniques capable of placement of optical nanoparticles with nanometer precision. Additionally, our strengths in single-nanoparticle spectroscopic characterization could be significantly enhanced by adding capability for single nano-element Raman and magneto-optical spectroscopy while expanding ultrafast capabilities to include single-photon counting 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 and pioneer the area of "phononics" (manipulation of phonons and phonon coupling phenomena).

## 2.3 Hierarchical structure & dynamics in soft matter



**Scientific opportunity:** A grand challenge in nanoscience integration is the ability to propagate the intrinsically unique behaviors of nanoscale materials into functional materials and systems at the macroscale. Nature provides a vast array of blueprints by which this challenge can be achieved in soft materials. For example, cephalopods (e.g., squids) are able to rapidly change their color at the organismal level based on changes in structure (and associated function) across multiple length scales, beginning with the active reorganization of pigment granules at the sub-cellular level. Our goal is to develop strategies (principles and soft materials) that will enable the hierarchical assembly of

individual nano-constituents so as to harness their collective or emergent behaviors. It is anticipated that the multiscale and multidimensional assembly of nanoscale building blocks will lead to next generation photonic (e.g., solid-state lighting, lasing, color tuning), electronic (i.e., beyond silicon electronics), and energy storage technologies.

**Positional science and capabilities:** Current CINT capabilities include the synthesis of a wide range of both natural and engineered functional nano-constituents, structured soft materials that can serve as platforms for the spatial/orientational organization of the individual nanocomponents, and multiscale modeling and visualization tools, all of which serve to position CINT for realizing this scientific opportunity. Specifically, CINT has established expertise in the large-scale production of naturally-derived, functional biomolecules including transport nanomotors, (kinesin and dynein motor proteins), light-driven proton pumps (bacteriorhodopsin), light-gated ion transporters (channelrhodopsin), and rotary actuators ( $F_1$ -ATP synthase). This effort is complimented by unique capabilities in the synthesis of engineered nanoparticles, including fluorescent metal nanoclusters, non-blinking quantum dots, high explosive detonation nanocarbons, plasmonic nanoparticles, magnetic nanoparticles, and chemically tailored carbon nanotubes and graphene.

In the area of hierarchically-structured soft materials for the assembly of nano-constituents into functional complexes / composites our capabilities include the development of bio-derived (lipid) matrices, artificial biomembranes, genetically-engineered responsive peptides / polymers, wholly synthetic block copolymer constructs (vesicles), structured dual conducting poly(ionic liquids), and stimuli-responsive blends of lipids and polymers.

Advanced characterization techniques are crucial for understanding the structure and functional dynamics of assemblies/complexes across a broad range of time and length scales. CINT has world-leading capabilities for the visualization of both the spatial distribution of nano-constituents within complex environments and their dynamics. Three-dimensional single molecule / particle tracking microscope, developed at CINT, images the fine details of complex dynamic systems, for example, the precise step sizes of motor proteins propagating within a biomembrane have been imaged (Figure 6). Super resolution fluorescence microscopy provides images of fluorescently labeled samples to a resolution of 10 ~ 20 nm (a factor of 10 below the diffraction limit of 250 nm), approaching the resolution of electron microscopy. In concert with these imaging techniques, correlated AFM and fluorescence imaging affords unique capabilities combining single-molecule sensitivity with time-correlated single-photon counting. These world-class visualization capabilities are augmented with an environmental scanning force microscope that yields information on the adhesion and binding characteristics of nanocomponents; and an in-house X-ray scattering (SAXS/WAXS) instrument which provides greater structural details (from 100's of nm to Å).

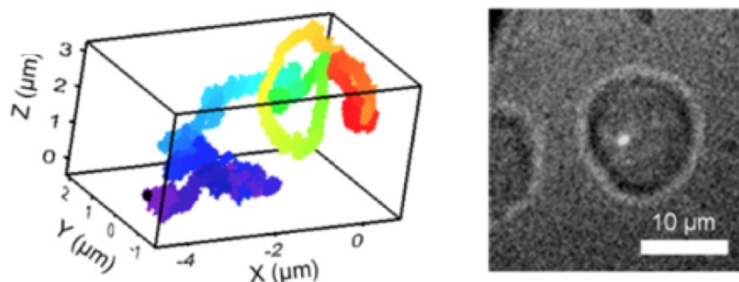


Figure 6. (Left panel) 3D trajectory of a single allergy antibody (IgE) labeled with a single gQD on a plasma membrane. (Right panel) White light image of the cell during the trajectory map. Ref. Keller et al. 2014 Adv. Func. Matter.

**Toward the future: Synthesis and fabrication of soft materials:** Drawing on the above mentioned strengths of CINT, we will push the state-of-the-nanoscience beyond the synthesis and fabrication of simple homogeneous building blocks (nano-constituents) and composites and seek to create integrated systems which combine multiple functional components and exhibit emergent properties

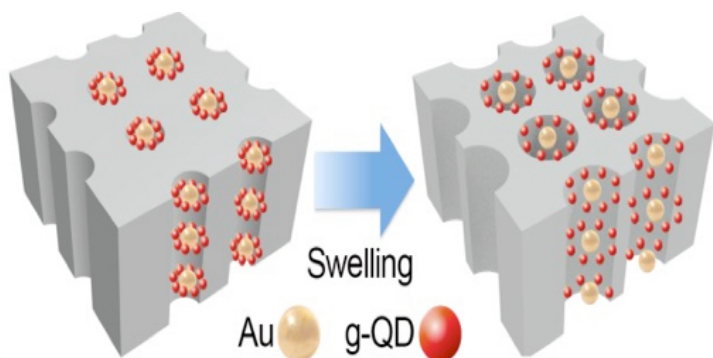


Figure 7. Water-induced swelling and contraction of a tetragonally perforated lamellar structured polymer serves to regulate macroscopic optical response of spatially organized plasmonic NPs (Au) and emitters (gQDs).

assembly of nanoparticles. By controlling the spatial arrangement of the individual nanocomponents within a responsive matrix will allow for dynamic tuning of their spatial proximity and therefore active regulation of the macroscopic properties of the material. For example, the spatial organization of nanoscale emitters (gQDs / nanocarbons) and plasmonic (metal) nanoparticles doped or *in situ* synthesized within a hierarchically-structured soft matrix will offer a means for achieving super-radiance, plasmon assisted lasing and dynamic control over photon emission or light interaction (“color tuning”), all component materials that could ultimately be integrated to form next generation

(Figure 7). In addition, the responsive soft matrices will promote dynamic and/or programmable interfaces that drive structural reorganization and associated changes in macroscopically observed composite/system function. Through optimization of our “toolbox” of stimuli-responsive, structured soft materials we will achieve multi-scale assembly of disparate nano-objects, serving to fulfill, for example, our vision of hybrid materials for the controlled manipulation of light (see section 2.2). Critical to achieving these goals will be the nano- to mesoscale



nanophotonic devices. Our current collection of structured soft materials, however, will require synthetic modification so as to possess the following materials attributes:

- Distinct regions for spatial organization of the functional nano-constituents,
- Environmental-responsivity for active reconfiguration of the nanoparticle arrangement and hence composite properties,
- Process-ability for extension of structure into macroscopic dimensions,
- Compliant interfacial chemistry for coupling to traditional device materials (e.g., metals and ceramics) / architectures,
- Requisite balance of mechanical durability without sacrificing dynamics.

Another area requiring significant investment will be infrastructure (instrumentation, tools, clean room space, and expertise) to expand our efforts in top-down patterning / lithography and processing of self-assembled (bottom-up) soft materials. That is, full realization of nanocomposites with complex functionality will require the ability to generate 2D and 3D patterned materials that will allow for the integration of nano-constituents over a full range of length scales (spanning nano → macro), this aspect of our work will require the addition of a Soft Fabrication Laboratory at CINT. The facility will contain a suite of 2D and 3D patterning tools, including capabilities for micro/nano-contact printing, molding, inkjet printing, optical lithography, and scanning-probe based direct write techniques.

*Strategic plan and investment:* Concurrent with advancing the fabrication of soft materials, substantial improvements in modeling tools applicable for understanding multi-length scale *and* temporal phenomena will also be pursued, aiding in the *a priori* design of the materials and analysis of multimodal characterization data. For example, LAMMPS codes development at Sandia will be used to improve our image analysis capabilities.

In recent years, considerable progress in optical microscopy has enabled characterization of soft and biological materials with spatial resolution well below optical diffraction limits, and with high temporal resolution. CINT has led the effort in developing imaging and visualization tools for single bio-macromolecule / particle tracking. While these techniques are well suited for evaluating the dynamics of individual nanoscale components, they often lack the ability to capture details of the molecular structure of complex assemblies. A full understanding of complex composite materials requires imaging structure and structural dynamics over a full range of length and time scales. Future efforts within CINT will focus on achieving this goal by improving optical tracking capabilities to achieve single molecule resolution and by combining other characterization tools, such as force microscopy, X-ray scattering and electron microscopy into a single platform. A specific opportunity for advancing multimodal characterization will be the coupling of X-ray scattering/ diffraction which provides atomic resolution with the images and trajectory information provided by 3D optical tracking. It is anticipated that integration of these techniques onto a single platform will yield structure and structural dynamics over the Angstrom to micron length scale.

*Meeting the modeling challenge:* CINT's ultimate goal in this area is to develop a fundamental understanding of the interactions among nanoscale components, and how these interactions affect the overall functionality of the hierarchical structures, thus leading to optimization and control by design.



The world leading LAMMPS molecular dynamics code is our base for performing these simulations. CINT will pursue the following directions:

- Develop exascale code capability to treat the hierarchical systems studied by the experimentalists through code development to use accelerators (e.g. GPUs) of exascale computers.
- Develop coarse-grained models from the underlying atomistic models to expand the spatial and temporal ranges significantly, while maintaining key (molecular) chemistry.

### 3.0 Scientific Expertise to Realize the Vision

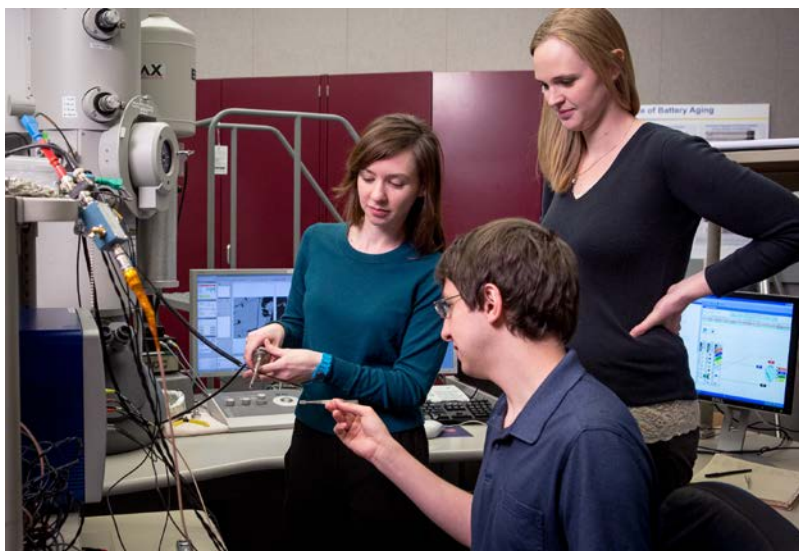


Nanomaterials integration involves:

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 1 (namely Nanophotonics & Optical Nanomaterials, Nanoscale Electronics & Mechanics, Soft, Biological & Composite Nanomaterials, and Theory & Simulation of Nanoscale Phenomena) 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 not only to retain our current workforce to maintain our forefront role in 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 2 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 be actively engaging our user community through a variety of mechanisms to promote integration of nanomaterials as well as to address challenges in scientific frontiers through both forefront research and development of unique capabilities important for the future new frontier nanoscience and nano-manufacturing.

## 4.0 Transformational and Foundational CINT Capabilities

Through a combination research expertise, specialized capabilities, and essential foundational techniques, CINT enables our users to perform complete research projects 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.

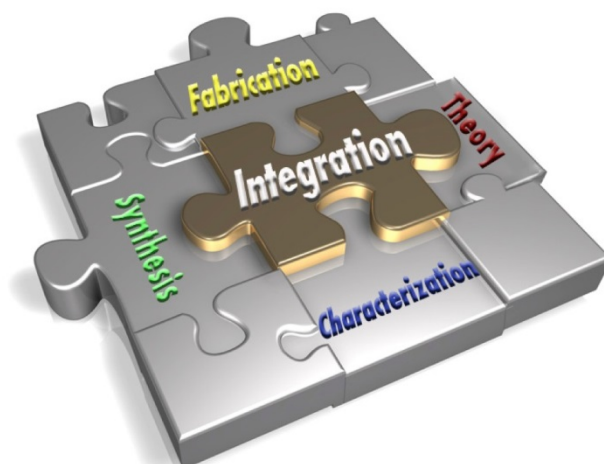


Figure 8. Addressing integration through collaborative capabilities and expertise in fabrication, synthesis, characterization and theory.

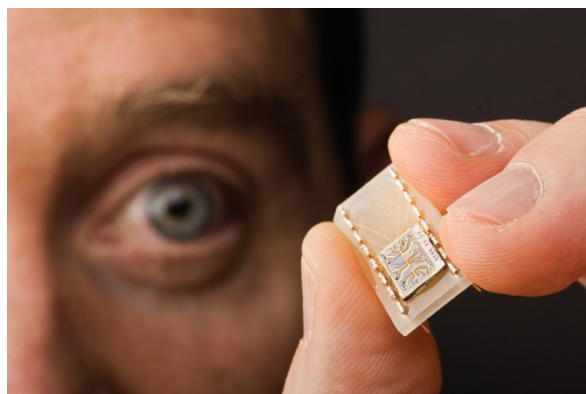
In this section, we identify a selection of CINT capabilities that will be important in addressing future integration challenges and that will transform the way integration research is conducted (CINT strategic objective 2). Some involve significant upgrades to current capabilities while others will 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](#). 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/SC user facilities is our portfolio of Discovery Platforms™.

### 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 the Microsystems Engineering Science and Applications (MESA) facility at Sandia National Laboratories.





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.

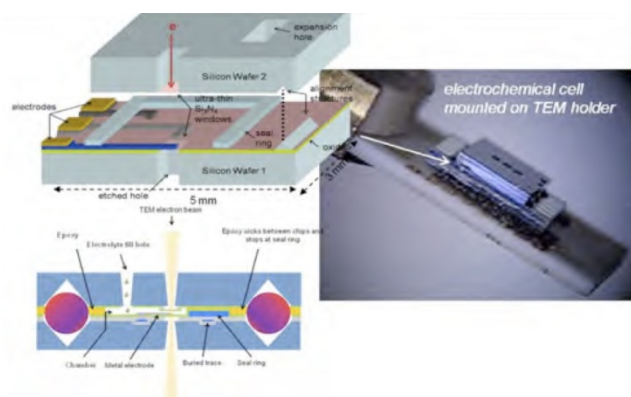


Figure 9. Electrochemical Discovery Platform.



Figure 10. Microfluidics Discovery Platform.

The present suite of Discovery Platforms includes:

**Electrochemical Discovery Platform:** The ElectroChem 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 9. This platform enables direct observations of solid/liquid interfacial process such as electrode/electrolyte interactions, electrode dissolution in electrolyte and solid-electrolyte interphase layer formation.

**Microfluidics Discovery Platform:** The new Microfluidic Synthesis Discovery Platform as shown in Figure 10 is an extremely flexible system for nanoparticle synthesis, functionalization, and real-time characterization. The microfluidic system utilizes an all-glass chip with a serpentine channel that can hold volumes from 200 microliters to one milliliter and precision temperature control. Real time reaction monitoring via visible and fluorescence microscopy, as well as UV-vis spectroscopy are currently being used with IR spectroscopy and dynamic light scattering to be added in the future.

The integration science challenges present exciting new opportunities to develop next-generation versions of the Discovery Platforms<sup>TM</sup> and to invent additional platforms in conjunction with CINT users designed for the synthesis and characterization of integrated nanomaterial systems (CINT strategic objective 3). As with all platform design cycles, the initial step is to specify the specific desired functions of a new platform, the performance criteria, and the scope of research for which it is expected to be used. These early platform concepts are then vetted with the external scientific community via Discovery Platform Workshops and focused sessions at the annual CINT User Meeting.

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 off of CINT's Microfluidic Discovery Platform<sup>TM</sup> 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 new development, there will be a strong emphasis on integrating the Microfluidics 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 and microfluidic delivery that will enable real-time control over multi-scale, hierarchical assembly.

An important new direction for CINT will be collaborations with other Nanoscale Science Research Centers and DOE/SC user facilities (light sources and neutron scattering centers) to couple the capabilities of a Discovery Platform<sup>TM</sup> with unique instrumentation at these other national user facilities. The future outcome will be not only an expanding portfolio of platforms optimized for nanomaterials integration at CINT but also a wider community of researchers using these microscale instruments for other cutting edge research Nationwide.

## 4.2 Synthesis / Fabrication

Creating novel 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:

**Semiconductor Molecular Beam Epitaxy (MBE):** It is in demand worldwide to grow high-purity, ultra-high mobility As-based III-V compound semiconductor structures with atomic monolayer precision for fundamental studies of 1-D and 2-D nanomaterials. Our future plan includes extending our MBE capabilities to group IV materials to provide users with a new family of 2-D layered materials.

**Heteroepitaxial Growth:** Nano-composite 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.

**Nanowires:** CINT specializes in the synthesis of semiconductor nanowires by solution-phase and chemical vapor deposition (CVD) approaches to produce single crystal nanowires, radial/axial heterostructured nanowires, and complex architectures consisting of Si/Ge, III-V and other compound semiconductor materials. Future directions include using microfluidic methods to have dynamic control and *in situ* monitoring during Flow-Solution-Liquid-Solid growth.

**Atomic-Precision Lithography:** One of our newest capabilities is Atomic-Precision Lithography, which involves the placement of phosphorus donor atoms into silicon with atomic-scale precision by hydrogen resist lithography in combination with scanning tunneling microscopy. *CINT is one of only two institutions in the world with this capability.*

**Integration Laboratory:** This fabrication laboratory is class 1000 cleanroom with a comprehensive suite of micro/nanoscale tools including Atomic Layer Deposition (ALD), Low Pressure CVD, Physical Vapor Deposition, Electron Beam and Photo Lithography, 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 current generation commercial tools.



Figure 11. Atomic Precision Lithography.

**Solution Phase Synthesis of Optical Nanomaterials by Design:** CINT is pioneering solution-phase approaches for generation of complex optical materials including routes to introduce anisotropic composition, structure, shape and function. Our fully software controlled automated parallel nano-reactor systems and in-flow microfluidic nanowire syntheses gives unprecedented control of timed nucleation and growth processes. By accessing through these methods a full compositional and interfacial parameter space we have developed non-blinking nanocrystal quantum dots that emit in the visible and the near-infrared. Microfluidic control of nanowire synthesis provides strategies to produce complex axially heterostructured nanowires with ultra-sharp interface regions.

**Chemical Vapor Deposition Capability for Growth of Semiconducting Nanowires and Thin Films:** Our 3" 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:** Development of carbon nanotube-based materials is enabled by CINT 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 understanding of separations mechanisms. We are pioneering low-level covalent doping strategies via solution and solid-state methods for 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 metamaterial, nanoresonator and plasmonic 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 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 in 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):** As a scanning probe lithographic technique, CINT DPN capability is highly complementary to other lithographic approaches for materials integration and heterostructure generation. Our expertise allows delivery of a broad range of deposition inks onto any desired substrate with precise positioning in the nanometer range. Our focus is on delivery of sub-10 nm to 50 nm sized nanocrystals, with an ability to write to 1-D, 2-D, and 3-D prepatterned structures of both soft and hard materials. Integration of optical emitters to dielectric optical antennae, plasmonic structures, and metamaterial assemblies has been demonstrated.

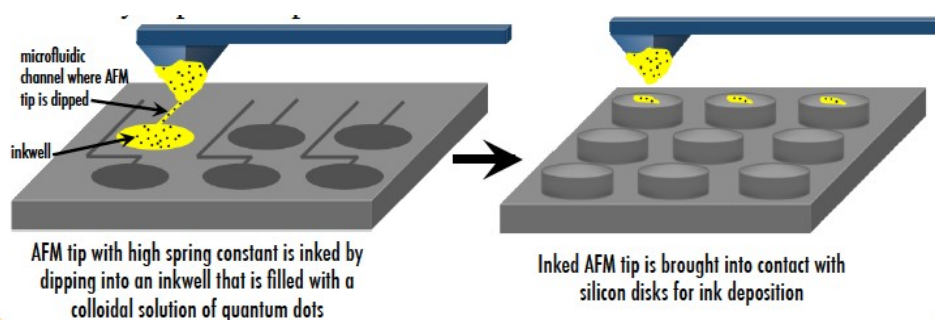


Figure 12. Dip Pen Nanolithography using a quantum dot ink.

**Integrated Quantum Photonic and Optomechanical Circuits:** Newly introduced to CINT is capability for development and application of photonic and phononic circuits. Integrated photonics and optomechanical capabilities at CINT include fabrication of photonic waveguide circuits, resonators, filters, beamsplitters and other optical and mechanical elements. Advanced optical packaging for on-off chip coupling of less than 0.5 dB per facet along with a suite of lasers from 765 nm to 1640 nm allow for a diversity of fundamental and applied research. Integrated photonic and phononic devices are being used in CINT for coherent classical and quantum information processing, wavelength conversion, entangled photon generation, quantum communications and computation, as well as for studying phonon-mediated optical squeezing, single-photon optomechanics, and related phenomena in individual nanostructures.

**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.

**Expression, 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.

**Genetically-encoded synthesis of fluorescent metal nanoclusters:** Novel metal nanoclusters that exhibit unique fluorescence, electronic and catalytic properties.

**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, robust polymer networks.

### 4.3 Characterization

CINT 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. Development of advanced probes plays a critical role in high-impact nanoscience discoveries and innovation of next generation technologies.

**Transmission Electron Microscopy:** 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 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; adding aberration-corrected TEM to the user program to remain internationally competitive; offering a new suite of *operando* capabilities as identified in the recent DOE workshop on the future of electron scattering; and developing the expertise/techniques for soft/composite nanomaterials microscopy needed by the integration challenges in section 2.

**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. Future plans for this platform include adding integrated lithographically defined heaters and mechanical actuation stages.

**Nanomechanics:** CINT is currently the only NSRC with a dedicated nanomechanics capability, which includes *ex situ* nanoindentation and electrical contact resistance measurements, integrated Atomic Force Microscopy, and oscillatory load cells. These are complimented by *in situ* straining stages for the Scanning Electron Microscope (SEM) and TEM.

**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. Photoelectron microscopy and dynamic experiments are enabled by the addition of deep-UV pulsed laser sources.

**Ultrafast Optical Spectroscopy:** It offers unmatched ability to differentiate the dynamics of spin, charge, and lattice, and coupling between them in time and spectral domains with femtosecond temporal resolution. CINT has a full range of time-integrated and time-resolved optical tools, covering the terahertz (THz) 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 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 4K 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 Si nanowires, ultrafast phase transition in VO<sub>2</sub> nanoparticles, and ultra-fast switching of metamaterials, to name a few.

**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 capability for controlling temperature (4-450K) and gas and humidity of the sample environment. Time correlated single photon counting capability also provides time-resolved PL and photon correlation/cross-correlation measurements using Silicon 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 ~ 2ps) and a fluorescence up-conversion system (temporal resolution ~150fsec) are also available for time-resolved PL measurements.

**Fully-Tunable Raman Spectroscopy and Microscopy:** CINT resonance Raman instrumentation provides 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 tune-ability from 345 nm to 1000nm. Among the many materials studied with Raman at CINT, examples include carbon nanotubes, graphene and other 2-D materials, SERS-active structures, bio and soft-material composites, quantum dots, nanowires, and multiferroic complex oxides.

**3-D tracking microscope:** Unique spatial filter geometry and active feedback in XYZ allows tracking of 3D motion of selected fluorophores, such as quantum dots, fluorescent proteins, or organic dyes moving at biologically relevant transport rates( $\mu$ ).

**Bessel beam plane illumination microscopy:** An alternative to conventional laser scanning confocal microscopy that enables rapid (100 frames per second) 3D imaging. 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  $\sim 10$ - $20$  nm, approximately factor of ten below the diffraction limit ( $\sim 250$  nm) and approaching that of electron microscopy (EM).

**Scanning probe spatially correlated atomic force microscopy (AFM) and fluorescence imaging:** A unique 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 built up 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.

**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 allows a spatial resolution of 10-20 nm throughout the electromagnetic spectrum from far infrared (THz) to visible. Information of local optical properties can be obtained by 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 with full-access of spectroscopy information, particularly in the mid-infrared wavelengths ranging from 4.7 to 15.3  $\mu$ m, and the terahertz (THz) frequency range (0.3-3 THz) to be integrated. The a-SNOM also includes pump-probe capability employing two femtosecond laser

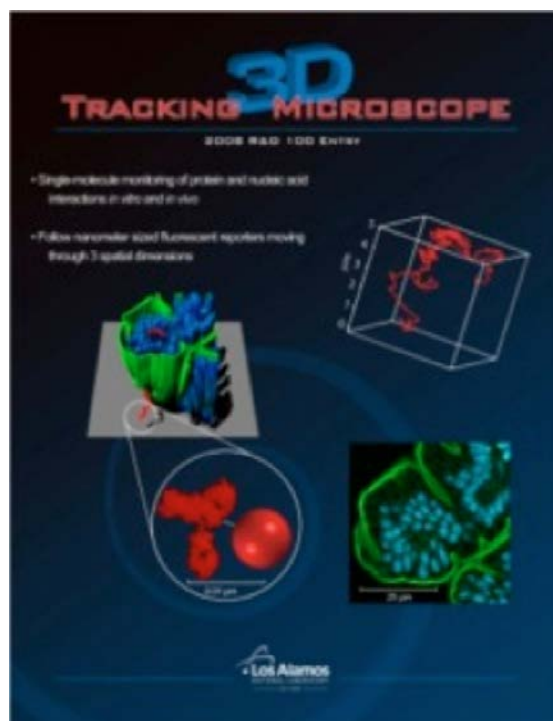


Figure 13. R&D 100 award winning 3D Tracking Microscope.



Figure 14. neaSPEC GmbH apertureless scanning near field microscope.

sources at 1560 nm and 780 nm. The capability is further enhanced by expertise provided by personnel in the NPON thrust and the Laboratory for Ultrafast Materials and Optical Sciences (LUMOS Team). Thus 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 nano-structured functional photonic devices.

**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 Modeling

The unique properties of nanostructured 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 component itself but also for its interactions with surrounding components and materials. In addition to foundational capabilities like density functional theory (DFT) for electronic and 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 (NA-ESMD) 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 NA-ESMD methodology offers a computationally tractable route for simulating hundreds of atoms on ~10 ps time scales where multiple coupled excited states are involved.

**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

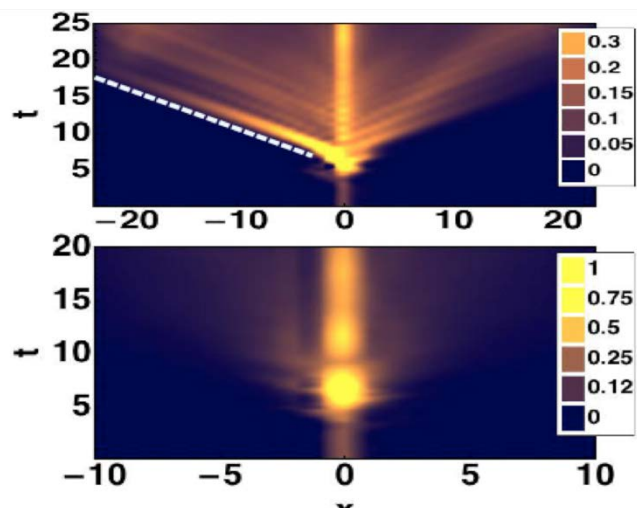


Figure 15. A polaron quasiparticle (an electron dressed by fully quantum phonons) in a nanowire is excited by an ultrafast optical pulse.

and effective modeling in complex materials. We will generalize this exact approach to treat the electronic correlation effects.

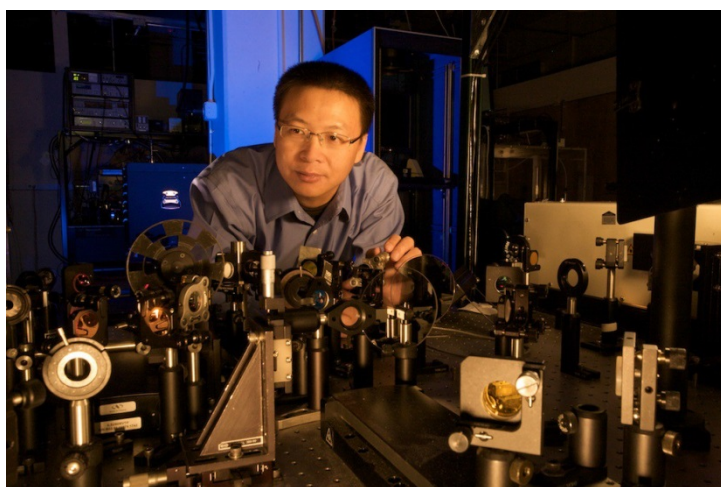
**Electronic Structure with Correlations:** (i) CINT is also pioneering the theoretical modeling of local electronic structure with strong correlation. The capability has been applied to understand the bulk properties of correlated electronic materials by studying the electronic signatures around local impurities and defects. We have also developed quantum many-body approaches (Quantum Monte Carlo and Gutzwiller Variational Wavefunction) 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. (ii) Although not leading, CINT has a track of record in applying the GW (“G” stands for the Green’s function and “W” for the screened Coulomb interaction) method to study the quasiparticle excitation properties in solids including actinides and 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. LAMMPS, the Sandia molecular dynamics code can treat both the nanoparticle core, the bound ligands and the surrounding liquid/polymer.

**Simulation in Energy and Time Domains:** We have developed the capability to analyze MD simulations in a static or oscillating electric field. In a similar vein, we can extract and analyze the dynamic structure factor from MD simulations to compare directly (in the energy domain) or indirectly (in the time domain) to quasi-elastic neutron scattering data.

## 4.5 Leveraging Capabilities

**Laboratory for Ultrafast Materials and Optical Science (LUMOS):** The LUMOS facility 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 terahertz-probe spectroscopy, high harmonic generation extreme ultraviolet spectroscopies, and scanning probe imaging and spectroscopies. We seek to understand and control the interaction of photons with materials’ electronic, spin, and structural behavior on the ultrafast time scale for missions of national security. To learn more about LUMOS capabilities, please visit <http://cint.lanl.gov/facilities/LUMOS/Capabilities.php>





**Ion Beam Materials Laboratory (IBML):** The core of the laboratory 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 research capabilities include routine ion beam analysis techniques such as Rutherford backscattering spectrometry (RBS), nuclear reaction analysis (NRA), elastic recoil detection (ERD), particle-induced i- 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. More information about IBML can be found at <http://www.lanl.gov/science-innovation/science-facilities/ion-beam-materials-lab/index.php>.



**Microsystems Engineering and Science Applications (MESA) Complex:** Sandia National Laboratories 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 is designed to 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 is 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. More information about MESA may be found at: <http://www.sandia.gov/mstc/>.

**In-situ Ion Irradiation Transmission Electron Microscopy (I<sup>3</sup>TEM) Facility:** The I<sup>3</sup>TEM facility (at SNL) 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 breath 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.

#### 4.6 Future Envisioned Capabilities

CINT is in the process of integrating a 9 Tesla superconducting magnet to our full range of excitation capability for performing single nano-element magneto-PL and magneto-Raman microscopy/spectroscopy. This capability will open a new area of CINT studies in magneto-induced and magneto-coupled optical behaviors as well as enabling novel Raman studies of a wide range of heterostructured optical, electronic, magnetic, and multiferroic materials.

CINT is planning for a significant expansion of our available excitation ranges for Raman spectroscopy. We envision adding a wavelength-doubled dye laser system to our current doubled-Ti:Sapphire excitation source to push excitations to wavelengths as short as 270 nm. The extended UV excitations will enable us to probe currently inaccessible electronic transitions and energy ranges and expand the set of materials types we can study.

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 optomechanical and heterostructured materials.

Expansion of single photon detection capabilities to the mid-IR is under construction using a combination of nonlinear upconversion techniques. CINT will evaluate the procurement of single-photon superconducting nanowire detectors operating at longer wavelengths if they become available.

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.

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 Sandia's parallel molecular dynamics code LAMMPS to run on GPUs and the new high performance architectures will provide unprecedented capability to model soft materials.

## 5.0 Engaging the National/International Scientific Community

Realization of our vision of nanoscience integration will continue to require the active engagement of our user community through a variety of mechanisms that both promote the essential practices of integrating nanomaterials and serve to build robust communities around key integrations challenges that by their very breadth and complexity transcend individual project/investigator efforts.



One of CINT's main goals is to attract research leaders in diverse institutions, early career scientists, and innovators in nanotechnology companies to create an expanded user community. To achieve this, it is necessary to target outreach activities that would increase awareness of our existing portfolio and capabilities within this identified group. We must have continued presence in high-profile journals, organizing symposia at national meetings and targeted workshops on nanomaterials integration challenges, hosting high-profile seminar series, and demonstrating the full suite of CINT capabilities and how users can utilize our expertise within all aspects of integration. We plan to take advantage of the [NSRC Portal](#) as one way for users to identify CINT capabilities and expertise.

CINT's unique and distinguished capabilities and expertise are already attracting a collection of experts and young scientists dedicated to solving challenges in nanoscience and nanotechnology. Through these networks, we have begun, and will continue to develop collaborative communities of scientists, sharing ideas, and working together to speed up development of the technological innovations that will come from solving these challenges.

To best take advantage of the user facility model, we are currently working on the development of partner user agreements. These partner users would guarantee access in return for an investment in CINT. While investment in an instrument is most common, other forms of investment from a partner user could include intellectual investment or expertise shared with CINT staff and other users. Partner user agreements will assist us in remaining in the forefront of nanoscience, through the development of new, targeted capabilities.

The success of nanoscience integration will be in the ability to incorporate the basic science techniques within an engineering application. CINT is in a unique position to bring together the science and engineering expertise of LANL and SNL to expedite the development of nanomaterials integration. Our users and staff are already working with engineers at SNL to design and develop the Discovery Platforms. This is a partnership that will continue and broaden as nanoassemblies become included in chips and other electronic devices.

## 6.0 Operational Excellence

CINT has increasingly developed an identity that transcends Laboratory boundaries, striving for operational "best-in-class" performance at the Center. Efficiency is at the forefront of our business

operations focus. We have adopted and embedded standard project management principles and methodologies that continue to be applied throughout the Center to ensure operational efficiencies and impactful cost savings.

The realization and implementation of the online User Program Management System will allow us to improve business intelligence operations throughout the Center. This system allows for creating workplace efficiency by leveraging digital technology to decrease the amount of manual hours lost on easily automated tasks and enabling the support of an increased number of users. The tool will increase data accuracy and maintain a shared repository that will provide historical, current and predictive views. This data will allow CINT to track trends in the user program to allow for better decision-making in capability development and stay abreast of increasing science focus areas within our user communities.



both Los Alamos and Sandia national laboratories.

With the pool of dedicated scientists available through our Users Executive Committee, Science Advisory Committee and users in general, CINT is well poised to look for trends in nanomaterials integration across the nation and the world. Between these personal interactions, and the ability to see trends across the user program, CINT will be poised to respond to the needs of the nanoscience community. This information will help to fill gaps in available capabilities, both through the development of specialized instrumentation and in the collective expertise of CINT staff and user community. Strategic hires of scientists, technologists, and postdoctoral researchers will be based around the needs of the community and addressing integration grand challenges.

As with any business operation, CINT is constantly benchmarking best practices for improving on processes and procedures. We have the opportunity to lead the complex in improving cross-laboratory processes, in particular with a goal of creating a safety-training program that would be acknowledged at

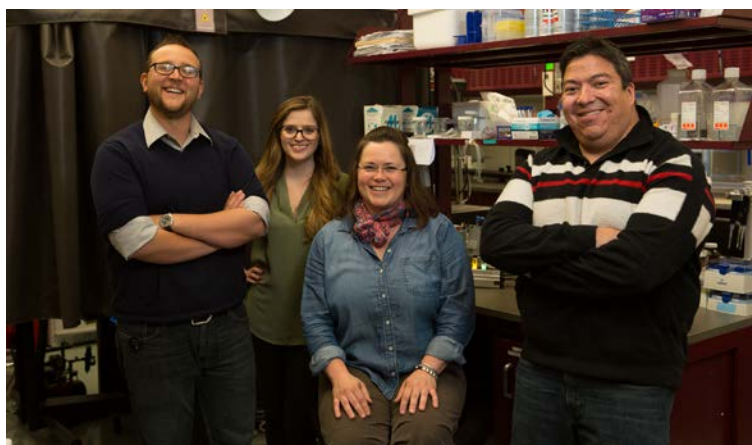


## 7.0 Implementation

CINT2022 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/SC user facility with a vibrant, growing user community, together position us to be a leader at this new frontier in nanoscience. Our six strategic objectives 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.



The 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 User Meeting, joint-NSRC workshops, and focused symposia at major national scientific conferences, consultation with the CINT Scientific Advisory Committee and the CINT Users Executive Committee.

When the National Nanotechnology Initiative was launched over a decade ago, the nation was at the dawn of the next technological revolution. Since then, amazing scientific discoveries have validated the promise that nanoscale is different. Just as integration transformed the transistor into the integrated circuit, now the scientific community is prepared to reveal even greater wonders 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.