AUTONOMOUS COMPUTING MATERIALS

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ABSTRACT

Conventional materials have reached their limits in computation, sensing, and data storage capabilities, ushered in by the end of Moore's Law in 2016, myriad sensing applications exceeding capabilities, and the continued exponential rise in worldwide data storage demand. Conventional materials are also limited by the controlled environments they must operate in, their high energy consumption, and their limited capacity to perform simultaneous, integrated sensing, computation, and data storage and retrieval. In contrast, the human brain is capable of multimodal sensing, complex computation, and both short- and long-term data storage simultaneously, with near instantaneous rate of recall, seamless integration, and minimal energy consumption. Motivated by the brain, and the need for revolutionary new computing materials, we recently proposed the data-driven materials discovery framework, *Autonomous Computing Materials*. This framework aims to mimic the brain's capabilities for integrated sensing, computation, and data storage by programming excitonic, phononic, photonic, and dynamic structural nanoscale materials, without attempting to mimic the unknown, implementational details of the brain. If realized, such materials would offer transformative opportunities for distributed, multimodal sensing, computation, and data storage in an integrated manner in biological and other, nonconventional environments, including interfacing with biological sensors and computers such as the brain itself.

INTRODUCTION

Conventional computing is founded on Boolean logic gates implemented in silicon-based hardware devices, which have emerged in the 21st Century after decades of research and development as the predominant computing materials platform. However, the recent end of Moore's law in 2016[1-2], the explosion of worldwide data usage ushered in by handheld and distributed devices, and emergent needs for new forms of biologically compatible, integrated sensing, computing, and data storage devices that have dramatically reduced energy footprints call for disruptive, radically new materials and substrates for sensing, computing, and data storage, beyond conventional silicon[3].

Specifically, the world's information is growing exponentially, from 10ZB today to an upper estimated limit of 1YB in 2030[4], with no practical materials solutions for long-term data storage in a low cost, energy efficient, archival manner. This growth is driven partly by the life sciences (genomics, proteomics, etc.); social media (Facebook, Twitter, Instagram, etc.); climatology, ecology, and cosmology; medicine and pathology; and finance, amongst other leading and growing applications. Increasing computational demand goes hand-in-hand with increasing data, driven by rapidly expanding applications of Artificial Intelligence and Machine Learning in all areas of work and life[5]. And sensing applications from autonomous cars and drones to wearables require orders of magnitude improvements in photonic, acoustic, and electronic spatial-temporal detection and sensing capabilities in order to operate reliably and safely. Finally, deployment of powerful computing and exquisite sensing capabilities are needed within living organisms, from agriculture to medical implants and wearables, as well as in resource-limited and harsh environments, with capabilities to interface with multiple modalities ranging from electronic to photonic and phononic.

While the human brain has long stood as a hallmark example of unprecedented natural computing, sensing, and storage capabilities that meet many of the preceding requirements, with numerous material modalities emulating neuronal networks and neuronal properties[6], only recently has our capability to perform large-scale recording of the spatial-temporal dynamics of the brain in complex environments involving decision-based learning through reward arisen. The brain's unique capabilities depend on the coordinated activity of numerous neurons (~100 billion in the human brain) forming a complex network. Thus, simultaneous recording of a large neuronal population is essential for understanding the emergent relationships among neurons and how they work together to perform their integrated sensing, computing, and information storage and retrieval functions. These techniques for recording brain activity *in vivo* now offer new opportunities to understand principles of the brain's operation, as well as to encode neuronal network data, computing/decision-making, and sensing properties into diverse materials. At the same time, control over material compositions, properties, and measurement modalities are offering new opportunities to program neuronal function, properties, and data into diverse frameworks including phononic silicon properties, excitonic DNA-scaffolded quantum dot and fluorophore networks, and dynamic nanoparticle structural networks, amongst other examples (**Figure 1**).

Motivated by a five-day ideation workshop organized and hosted by the National Science Foundation in 2019, called *Harnessing the Data Revolution*, we established the overall premise of *Autonomous Computing Materials*. We hypothesized that state-of-the-art, large-scale neuronal recording datasets could be leveraged to discover new means of programming dynamic, hierarchical, spatial-temporal sensing, computing, and data-storage capabilities into programmable nanoscale materials. As a 10-year vision for the field, we postulated that these encoding principles could also be applied to diverse, heterogeneous and stochastic datasets ranging from ecology to climatology, finance, etc., using a range of dynamic, controllable material properties including photonic, excitonic, phononic, magnonic, and structural, amongst others.

Importantly, our application of biomimetic ACMs is distinct from the well-established field of neuromorphic computing[7]. There, synaptic structure is typically replicated or mapped onto a silicon-based architecture or field-programmable neural arrays[8]. And increasingly, other materials are being considered for the creation of networks that may be literal analogs of synaptic networks in the context of transistor-based architectures. Notwithstanding, that paradigm primarily aims to reconstruct the von Neumann architecture, whereas in an *Autonomous Computing Material* we are expressly trying to employ a neural computing architecture. That is, we posit the possibility of constructing self-contained ACMs consisting only of complex materials—e.g., DNA-based photonic and excitonic materials; engineered nanoparticles scaffolded within polymer network arrays; or silicon-based phononic materials—that allow for autonomous computing without reliance on, or connection with, the now standard architectures of silicon-based transistors, and with minimal energy requirements.

NEURAL RECORDING TECHNIQUES

The human brain is arguably one of the most sophisticated computational machines known to humankind, having evolved to achieve high levels of capacity and accessibility of data storage, as well as extreme versatility and flexibility in the type of computation performed. Since the introduction of the neuron doctrine by Santiago Ramon y Cajal in the late 19th century[9], it is appreciated that the brain consists of discrete units called neurons forming complex circuits. It is the flexible connections and coordinated activity of numerous neurons, approaching 100 billion in humans, that enable functions such as perception, cognition, and movement.

A typical approach aimed at understanding the relationship between the brain's activity and its function is to record the activity of neurons in behaving animals. Due to technical limitations, a traditional approach has been to insert a single metal electrode in the brain to record the activity of one or a few neurons at once. The low throughput of these experiments has to-date hindered our understanding of how the numerous neurons work together to mediate brain functions.

The last decade has seen a technical revolution in our ability to record larger numbers of neurons. Techniques include electrophysiological recordings with high-density electrode arrays[1-2] and optical imaging. In particular, calcium imaging has emerged as a powerful approach to optically record the activity of many neurons simultaneously in an intact brain with minimal invasiveness. With this technology, it is now possible for a single study to include recording from tens of thousands of neurons[3], several orders of magnitude improvement compared to more traditional approaches. Thanks to these new approaches, a wealth of data representing the ensemble brain activity is beginning to accumulate. This presents an opportunity to uncover ensemble and emergent properties of neural populations in the brain and understand and potentially utilize fundamental operational principles of the brain, as well as mimic or encode them into nanoscale materials.

Despite this opportunity, traditional means of analyzing neural data have largely focused on single neuron activity, and the descriptions of population dynamics and their operational principles are in their relative infancy. An important opportunity is to leverage emerging data science techniques to extract key features of high-dimensional brain activity.

In *Autonomous Computing Materials*, these preceding properties are being explored in diverse materials frameworks, including nanoscale photonic and excitonic networks programmed using DNA nanotechnology; phononic systems programmed in silicon-based materials; and dynamic structural networks of gold nanoparticles and quantum dots interacting through nucleic acid interactions (**Figure 1**). The over-arching aim is to realize novel materials frameworks that sense, compute, and store information in an integrated manner, similar to the brain, without relying on von Neumann architectures or literal implementational details of synaptic connectivity, as leveraged in the well-established field of neuromorphic engineering and computing per se.

DNA-BASED PHOTONIC AND EXCITONIC MATERIALS

Light can be controlled using structured DNA-based nanoscale materials in a variety of manners[10] ranging from organizing gold nanoparticle 2D and 3D organization and structure to control Raman scattering, plasmonic field enhancement[11-12]; circular dichroism and optical rotatory dispersion effects[13]; light diffraction for visible color control using crystalline structures with regular lattice spacing on the 100–600nm scale[14-15]; to organizing chromophores[16] and quantum dots[17] for exciton delocalization and transport on the nanoscale to mimic light harvesting systems[18].

In particular, scaffolded DNA origami[19] offers unprecedented nanoscale control over the asymmetric spatial positions of arbitrary numbers of secondary molecules including quantum dots and dyes to form discrete networks of interacting excitonic and photonic components[18,20]. While spatial chromophore organization has been utilized by plants and bacteria to harness sunlight for chemical energy, it has only recently begun to be explored in DNA-based materials to program integrated sensing, information storage, and computation[16,21-22].

In ACM, we are exploring how 1D, 2D, and 3D nanoscale spatial organization of quantum dots, dyes, and dye clusters can be used to mimic neuronal connectivity and network properties to encode Boolean logic, sensing, and decision-making. As a starting point, FRET-based networks are being used to emulate neuronal sensing, learning, and decision making from the mouse brain, based on recordings from the Komiyama lab[23] (**Figure 1**). As these DNA-based quantum dot and dye networks evolve in their theoretical underpinnings, and their capabilities are implemented experimentally, key questions we seek to explore include how to extract complex behavior and learning from large-scale *in vivo* neuronal recordings; how to encode behavior and learning within these networks; how to program robustness and error-correction that is a hallmark of the brain; how to integrate its multi-modal sensing, recording, and computational abilities; and how to facilitate these capabilities in an autonomous manner that consumes minimal-to-no-energy, while also operating in non-conventional wet, physiological, and other uncontrolled environments. In parallel, synergistic work, we are using DNA as an information-coding polymer itself[24], with a density that exceeds 10^{18} bits per cubic millimeter, random-access capabilities[25-26], and a shelf-life that can be extended to millenia using silica encapsulation[27].

DYNAMICAL NANOPARTICLE NETWORKS

In a complementary approach, we are exploring the use of dynamic networks of nanoparticles to mimic the brain. The neuronal network paradigm involves nodes that are linked together to a large number of nodes at varying distances. The degree of these nodes, the connectivity and the strength of the links can vary and in principle be reinforced through a learning process. To mimic this type of network with molecular materials, the nodes have to be large enough to accommodate such variable and reversible connectivity, and the links need to accommodate variable lengths. Engineered nanoparticles (ENPs) can be made large enough to provide a range of binding sites[4,28-29] and can be decorated to provide specificity[30-32]. Meanwhile, polymers of varying lengths can be used to provide physical binding to the ENPs and the links between them[33] (**Figure 1**).

To store and retrieve information, we postulate that we can either use a relational array of the degree of connectivity of a given set of nodes, or be process-driven by encoding information through its response to input signals. To compute, such networked ENPs would then need to accommodate signal transport, perhaps by replacing the linked polymers with ones that conduct. The nodes would then conduct the signals between the attached polymers as modulated by the extent of their respective binding. Such binding may indeed be enhanced or decreased by the strength of past signals—memory—and thus allow for the network ENP to be trained. The processing of information—that is, signals—through this array would then need to be understood or designed using the operating system of the neuron that we are teasing out of the neural mouse model. This is the sense in which an ACM is being designed to mimic the underlying operating system architecture of the brain, as opposed to a von Neumann computer.

A key question for the implementation of an ACM based on an ENP-network is the degree to which the spatial-temporal signals transport through them and whether they follow the same rules as seen in time-series neuronal signals. To this end, we are using very large data sets (comprised of multiple signals from many interconnected neurons over a long period of time during which a mouse is subject to varying stimuli) to design the basic connectivity and operations in the ENP networks.

PHONONIC ENSEMBLES ENCODED IN SILICON-BASED MATERIALS

Phonons, the quanta of lattice vibrations, play an increasingly important role in information-processing applications both directly and through interaction with electrons and photons[34]. Control over phonons therefore has major implications in microelectronics[35-39], renewable energy harvesting[40], optoelectronics[41], and quantum technologies[42-43]. Additionally, phonons couple distinct components in heterogeneous systems, providing a natural platform for information storage and transfer in computing materials. However, the role of phonons as information carriers is considerably less explored. One main reason is that phonons are bosons and as a consequence, a broad range of phonon frequencies are excited at room temperature in condensed systems. The difficulty of working with a broad spectrum naturally poses challenges to control phonons[44]. Recent advances of nanofabrication and characterization techniques demonstrated remarkable possibilities to engineer phonon processes with nanostructuring[45]. Phonons in nanostructured materials revealed dramatic changes in their dynamics due to confinement[46-48]. In ACM, we aim to harness emergent phononic properties for a new paradigm for information storage and transfer, alternative to conventional charge or spin based computing protocols. Specifically, we hypothesize that stimulus-response of phononic ensembles can be regulated to exhibit collective dynamics similar to neuronal activity encoded in neuroimaging data. Significant advances in current understanding of structure-processing-property relationships between nanoscale structures and phonon processes promise to help realize such an engineered ensemble.

As a proof of concept, in ACM, we develop a computational framework to characterize emergent phonon properties of silicon-based nanoscale confined materials, such as in a FinFET device (**Figure 1**). While silicon-based structures have been used for electronics, and opto-electronics, there is little knowledge available regarding their complex phononic ensemble. The framework will potentially reveal new physics such as the coexistence of particle- and wave-like phonon phenomena. Some key questions we aim to answer are: is there similarity between the stochastic nature of large-scale neuronal data and quantized vibrations of heterogeneous assembly of nanoscale materials; and how can we design materials with a targeted phononic environment that exhibits desired data structure properties that can mimic neuronal state transition behavior. Such a model potentially will uncover new explorable design degrees of freedom to control thermal environments of high-impact technological applications.

BRIDGING NEURONAL COMPUTING WITH SOLID-STATE MATERIALS USING DATA-DRIVEN DISCOVERY

In order to bridge the gap from raw, digital neuronal datasets to nanomaterial systems, close interaction is needed between materials scientists, neuroscientists, and data scientists. Toward this end, data analytics serves a crucial, central role as models of dynamic, spatial-temporal processes are captured and mapped onto physical, materials systems. While there are advantages of physically mimicking the brain as performed in neuromorphic computing, a distinguishing feature of the ACM framework is that we are instead seeking to identify and create abstractions of the brain's computing and storage paradigms when it engages in complex tasks such as learning, decision-making, and sensing.

As an example, consider the abstraction provided by the Turing Machine, which defines clearly the notions of "computability," "encodability," and "decidability." While this architecture is simple and equivalent to a von Neumann architecture, it is also sufficient to compute any "function". Motivated by this example, we posit that all complex phenomena can be described in terms of canonical program that delineates the minimal number of states and unambiguously describes the transitions between them. This canonical program will be learnt from fully and partially observable phenomena injecting a certain iota of uncertainty.

A practical analogy can be found when the canonical program is realized as a field programmable gate array (FPGAs) of any form factor and material. Next is to embed this FPGA into other systems namely ones that are excitonic/patchy nano particle systems/phonic systems. The mapping is non-trivial and will require the mapping of states and transitions to other physical systems. By doing this, we are going beyond the manner in which neuromorphic computing is practiced now, whereby an entire "program" is emulated in a similar way on a different system. Thus, instead of creating "lego-brains" and smart chips that emulate it, our goal is to use other configurations so provided by nature. Further, the disconnect between the computing and algorithmic substrates no longer exists. The algorithm will now drive the embedded material system directly. Toward this end, new rules of programming and controlling the target materials systems are needed, which can and likely will lead to new foundational discoveries of computing (**Figure 1**).

HARNESSING THE DATA REVOLUTION

In order to best enable progress along the highly interdisciplinary research lines outlined above that span neuroscience, data science, and distinct areas of materials science, we propose several recommendations for progress towards *Autonomous Computing Materials* in our collaborative framework (**Figure 2**). First, we should work towards a common vernacular to facilitate communication across diverse backgrounds, and training. To achieve this, graduate students, postdocs, and faculty involved must collaborate closely, in order to bridge highly inter-disciplinary spaces, communicate across boundaries between traditional disciplines, in order to define and evolve a new language or common vernacular for inter-disciplinary materials and computational science. Second, transferability across materials systems is required, so that discoveries and inventions made in one domain, such as photonics and DNA-based materials, networked ENPs, or phononics and silicon-based materials, may interchange readily. To achieve this, iteration between distinct materials domains is needed, as data-driven materials discovery frameworks are explored, invented, and deployed. In order to ensure transferability, it must be maintained as an over-arching design principle of data-driven discovery approaches. Third, applicability across distinct, large-scale, stochastic, heterogeneous data-sets is required, not only from brain science, but also climatology, ecology, biology, pathology, etc. Fourth, industrial and government stake-holders, including the public, must be identified in order to identify new datasets and materials to transfer and apply the preceding methodologies and knowledge frameworks to, for broadest impact of the ACM framework on Industries of the Future[49]. Regular workshops and reports, first launched by the NSF in Washington DC in April of 2019 and held again later in May 2020, and embodied in this communication, are several such examples. Broader impacts on the public must be identified and communicated clearly, from enabling next-generation sensing for safe and reliable autonomous vehicles to heart and brain monitors for health and disease monitoring and treatment, and ecological preservation to avert impacts of global warming, are several examples of such domains that stand to benefit by transformations in our ability to compute autonomously in diverse materials and environments, akin to the human brain.

EMERGENCE OF *AUTONOMOUS COMPUTING MATERIALS*

We now have the ability to design materials with tunable properties across a broad parameter space that is too large to explore without deep learning techniques. We can now begin to access the spatial-temporal responses of a living brain to determine how it processes and stores information. We understand how to encode instructions into Turing machines (and quantum computing architectures) in ways that allows us to move towards the biomimetic architecture of the brain. Placing these pieces together with the revolutionary tools of the data science revolution suggests the feasibility for the design of novel materials that can carry our computing tasks effected by the flow of energy or electrons through them in much the same way that data flows and processes in the brain. Such *Autonomous Computing* holds promise both for the construction of large high-performance computing devices, and small portable devices capable of being integrated into fabrics and products. Here we have provided a preview for the underlying science of these devices and the possible technology that it will enable. In a timely quote by R.S. Williams from Hewlett-Packard Labs in 2017[3], "*The end of Moore's law may be the best thing that has happened in computing since the beginning of Moore's law. Confronting the end of an epoch should enable a new era of creativity by encouraging computer scientists to invent new biologically inspired paradigms, implemented on emerging architectures, with hybrid circuits and systems that combine the best of scaled silicon CMOS with new devices, physical interactions and materials.*" Indeed, *Autonomous Computing Materials* is one response to this challenge.

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FIGURES

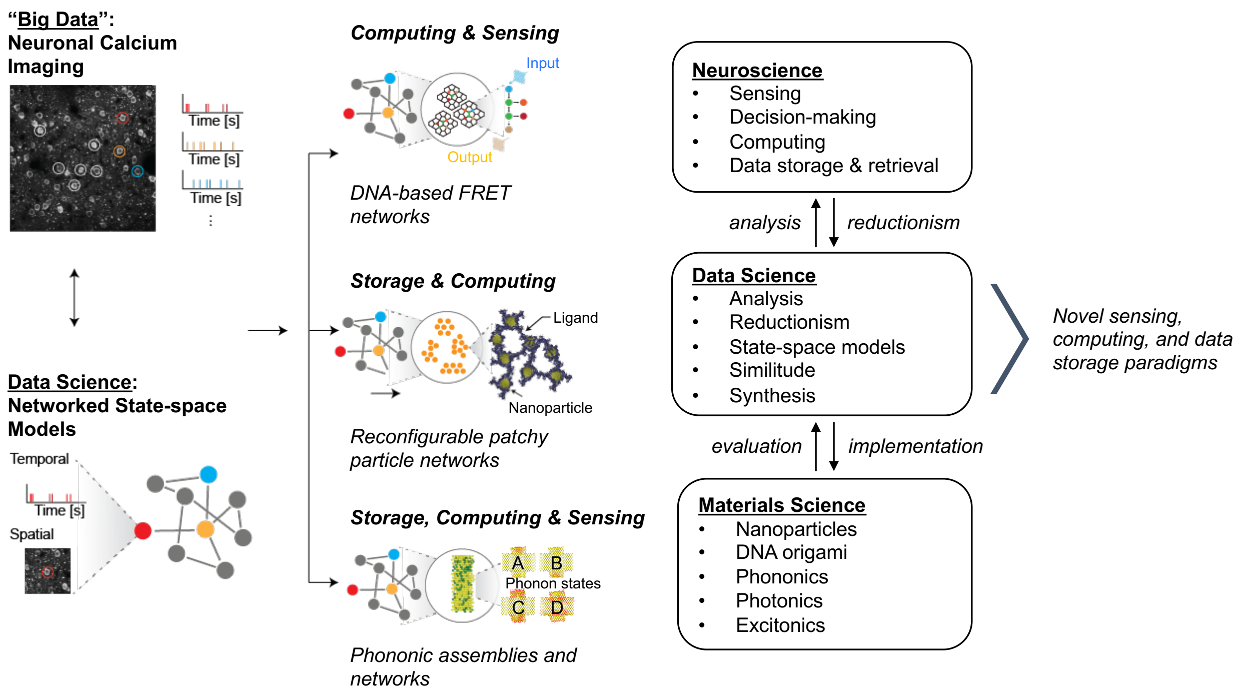

Figure 1. Investigative framework for discovering *Autonomous Computing Materials*, showing domain-specific opportunities and approaches associated with neuroscience, data science, and materials science.

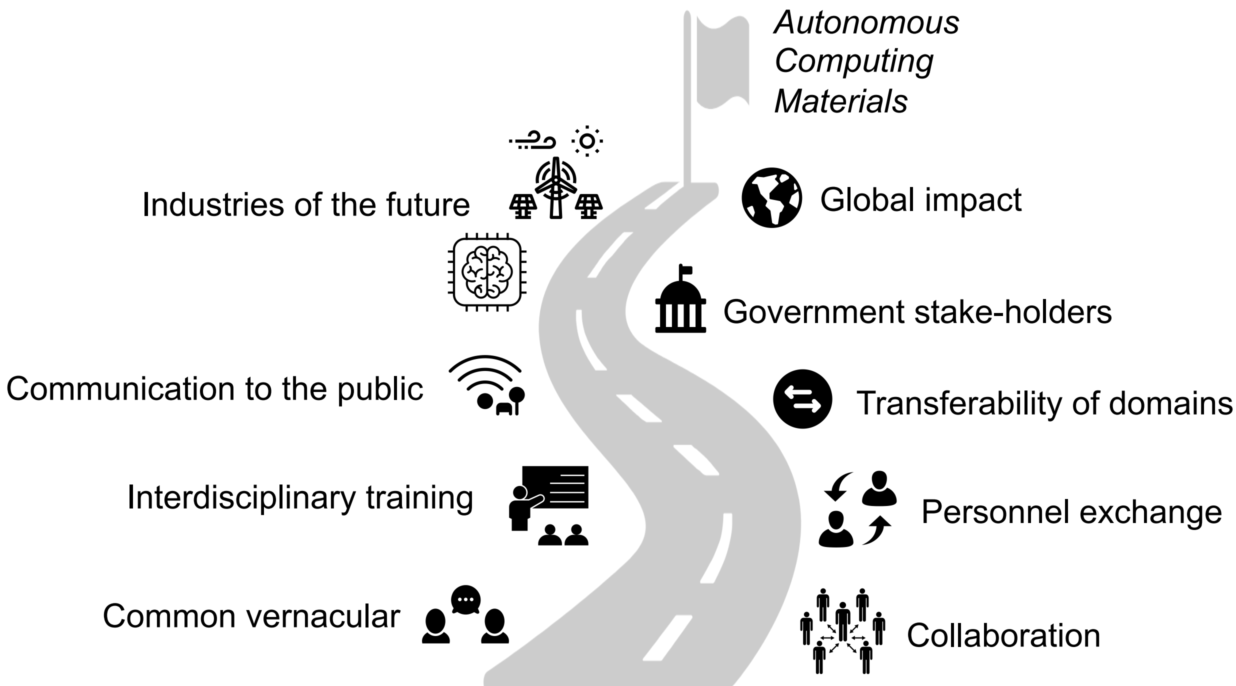

Figure 2. Proposed collaborative, inter-disciplinary strategy to enable the integration of neuroscience, data science, and materials science to discover and disseminate new modes of data storage, sensing, and computing with *Autonomous Computing Materials*.

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