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PERSPECTIVE

Autonomous Computing Materials

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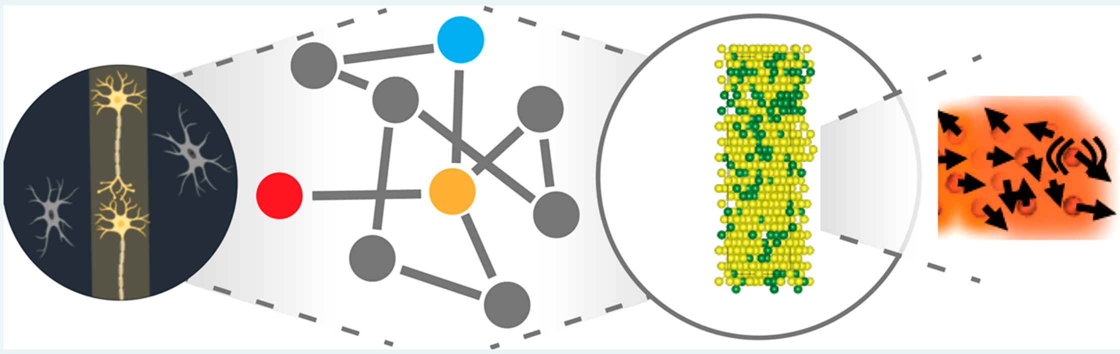

ABSTRACT: **Conventional materials are reaching their limits in computation, sensing, and data storage capabilities, ushered in by the end of Moore's law, myriad sensing applications, and the continuing exponential rise in worldwide data storage demand. Conventional materials are also limited by the controlled environments in which they must operate, 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.**

Conventional computing is founded on Boolean logic gates implemented in silicon-based hardware devices, which, after decades of research and development, have emerged 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 hand-held and distributed devices, and emergent needs for new forms of biologically compatible, integrated sensing, computing, and data storage devices with minimal energy footprints call for disruptive, radically new materials and substrates for sensing, computing, and data storage.[3]

The world's information is growing exponentially, from 10 ZB today to an upper estimated limit of 1 YB in 2030,[4] with no practical economical, energy-efficient materials solutions for long-term archival data storage. This growth is driven by the life sciences (genomics, proteomics, *etc.*); social media (Facebook, Twitter, Instagram, *etc.*); climatology, ecology, and cosmology; medicine and pathology; and finance, among 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] In addition, 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, powerful computing and sensing capabilities that can interface with multiple modalities including electronic,

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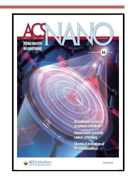

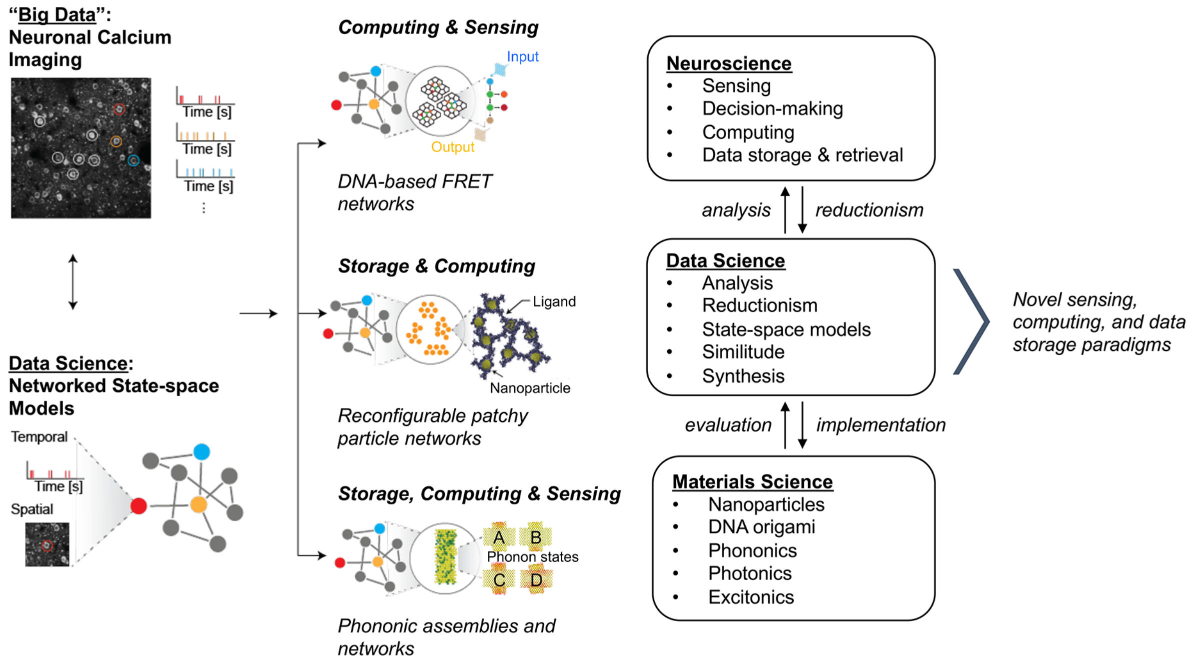

Figure 1. Investigative framework for discovering autonomous computing materials, showing domain-specific opportunities and approaches associated with neuroscience, data science, and materials science.

photonic, and phononic are needed both within living organisms, with applications ranging from agriculture to medical implants and wearables, and in resource-limited and harsh environments.

The human brain has long stood as the hallmark example of unprecedented natural computing, sensing, and storage capabilities. Although numerous material modalities emulate neuronal networks and neuronal properties,[6] our ability to perform large-scale recording of the spatial-temporal dynamics of the brain in complex environments involving decision-based learning through reward has only recently arisen. The brain's unique capabilities depend on the coordinated activity of numerous neurons (~100 billion in the human brain) that form a complex network. Thus, simultaneous recording of large neuronal populations 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.[7] These techniques for recording brain activity *in vivo* offer 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 offers 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 (NP) structural networks, among 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 (ACMs). We hypothesized that state-of-the-art, large-scale neuronal recording data sets 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 data sets ranging from ecology to climatology, finance, *etc.*, using a range of dynamic, controllable material properties including photonic, excitonic, phononic, magnonic, and structural properties, among others.

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Importantly, our application of biomimetic ACMs is distinct from the well-established field of neuromorphic computing.[8,9] There, synaptic structure is typically replicated or mapped onto a silicon-based architecture or field-programmable neural arrays.[10] Increasingly, other materials are being considered for the creation of networks that may be literal analogues of synaptic networks in the context of transistor-based architectures. Notwithstanding, that paradigm primarily aims to reconstruct the von Neumann architecture, whereas in an ACM, we are expressly trying to employ a neural computing architecture. We posit the possibility of constructing self-contained ACMs consisting only of complex materials—*e.g.*, DNA-based photonic and excitonic materials, engineered NPs scaffolded within polymer network arrays, or silicon-based phononic materials—that enable autonomous computing without reliance on, or connection with, the now standard architectures of silicon-based transistors and with minimal energy requirements.

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NEURAL RECORDING TECHNIQUES

The human brain is arguably one of the most sophisticated computational machines known, 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,[11] researchers have recognized that the brain consists of discrete units called neurons that form complex circuits. The flexible connections and coordinated activity of numerous neurons, approaching 100 billion in humans, 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 numerous neurons work together to mediate brain functions.

The past decade has seen a technical revolution in our ability to record larger numbers of neurons. Techniques now include electrophysiological recordings with high-density electrode arrays[1,2] and optical imaging. In particular, calcium imaging has emerged as a powerful approach to 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 recordings 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 ensemble brain activity is beginning to accumulate. These data present opportunities to uncover ensemble and emergent properties of neural populations in the brain, to understand and, potentially, to utilize fundamental operational principles of the brain, as well as to mimic or to encode these principles into nanoscale materials.

Despite these opportunities, traditional means of analyzing neural data have largely focused on single-neuron activity, and 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 ACMs, 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 (Au NPs) and quantum dots interacting through nucleic acid interactions (Figure 1). The overarching 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,[12] including using Au NP two-dimensional (2D) and three-dimensional (3D) organization and structure to control Raman scattering, plasmonic field enhancement,[13,14] circular dichroism and optical rotatory dispersion effects,[15] light diffraction for visible color control using crystalline structures with regular lattice spacings on the 100−600 nm scale,[16,17] and organizing chromophores[18−21] and quantum dots[22] for exciton delocalization and transport on the nanoscale to mimic light-harvesting systems.[23] In particular, scaffolded DNA origami[24] 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.[23,25] Although 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.[18−21,26]

With ACMs, we explore how the one-dimensional (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 (Figure 1).[27] Because 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, which are hallmarks of the brain; how to integrate multimodal 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 nonconventional wet, physiological, and other uncontrolled environments. In parallel, synergistic work, we are using DNA as an information-coding polymer itself,[28] with a density that exceeds 10^{18} bits per cubic millimeter, random-access capabilities,[29−31] and a shelf life that can be extended to millennia using silica encapsulation.[32]

DYNAMICAL NANOPARTICLE NETWORKS

In a complementary approach, we are exploring the use of dynamic networks of NPs to mimic the brain. The neuronal network paradigm involves a large number of nodes that are linked together 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 NPs (ENPs) can be made large enough to provide a range of binding sites[4,33,34] and can be decorated to provide specificity.[35−37] Meanwhile, polymers of varying lengths can be used to provide physical binding to the ENPs and the links between them (Figure 1).[38]

To store and to 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

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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, enable the network ENPs 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 template offers a means by which an ACM can be designed to mimic the underlying operating system architecture of the brain, as opposed to a von Neumann computer.

Key questions for the implementation of an ACM based on an ENP network include the degree to which spatial-temporal signals transport through the network and whether the signals follow the same rules as seen in time-series neuronal signals. To this end, we are using large data sets (comprising 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 increasingly important roles in information-processing applications both directly and through interactions with electrons and photons.[39] Control over phonons, therefore, has major implications in microelectronics,[40−44] renewable energy harvesting,[45] optoelectronics,[46] and quantum technologies.[47,48] In addition, phonons couple distinct components in heterogeneous systems, providing a natural platform for information storage and transfer in computing materials. However, the roles of phonons as information carriers are considerably less explored. One 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 in controlling phonons.[49] Recent advances in nanofabrication and characterization techniques demonstrated remarkable possibilities for engineering phonon processes with nanostructuring.[50] Phonons in nanostructured materials reveal dramatic changes in their dynamics due to confinement.[51−53] With ACMs, we aim to harness emergent phononic properties to create a new paradigm for information storage and transfer, alternative to conventional charge- or spin-based computing protocols. Specifically, we hypothesize that the stimulus response of phononic ensembles can be regulated to exhibit collective dynamics, similar to neuronal activity encoded in neuroimaging data. Significant advances in the 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, we are developing a computational framework to characterize emergent phonon properties of silicon-based nanoscale confined materials, such as in a FinFET device (Figure 1). Whereas silicon-based structures have been used for electronics and optoelectronics, there is little knowledge available regarding their complex phononic ensembles. 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 as follows: Is there similarity between the stochastic nature of large-scale neuronal data and quantized vibrations of heterogeneous assemblies of nanoscale materials, and how can we design materials with a targeted phononic environment that exhibits the desired data structure properties to mimic neuronal state transition behavior? Such a model will potentially uncover new explorable design degrees of freedom to control thermal environments in high-impact technological applications.

With autonomous computing materials, we aim to harness emergent phononic properties to create a new paradigm for information storage and transfer, alternative to conventional charge- or spin-based computing protocols.

BRIDGING NEURONAL COMPUTING WITH NANOMATERIALS USING DATA-DRIVEN DISCOVERY

To bridge the gap from raw, digital neuronal data sets to nanomaterial systems, close interaction is needed between materials scientists, neuroscientists, and data scientists. Toward this end, data analytics serves a critical, central role as models of dynamic, spatial-temporal processes are captured and mapped onto physical, materials systems. Although 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 to 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 the notions of "computability", "encodability", and "decidability". Although 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 learned from fully and partially observable phenomena injecting an iota of uncertainty.

A practical analogy can be found when the canonical program is realized as a field programmable gate array (FPGA) of any form factor and material. This FPGA is embedded into other systems, namely, ones that are excitonic/patchy NP/phononic systems. The mapping is nontrivial and will require the mapping of states and transitions to other physical systems. In so doing, we are going beyond the manner in which neuromorphic computing is typically 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 them, our goal is to use other configurations, as provided by nature. Further, the disconnect between the computing and algorithmic substrates no longer exists. The algorithm will now drive the embedded materials systems 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 in computing (Figure 1).

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HARNESSING THE DATA REVOLUTION

In order to 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 toward ACMs in our collaborative framework (Figure 2). First, we should work toward a common vernacular to facilitate communication across diverse backgrounds and training. To achieve this commonality, graduate students, postdocs, and faculty must collaborate closely, bridging highly interdisciplinary spaces and communicating across boundaries between traditional disciplines, in order to define and evolve a new language or common vernacular for interdisciplinary materials and computational science. Diversity should be encouraged in student, staff, and faculty backgrounds, training, genders, and ethnicities, to ensure creative and productive teams.[54,55] Second, transferability across material 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. As data-driven materials discovery frameworks are explored, invented, and deployed, iteration between distinct materials domains is needed to achieve transferability. In addition, transferability must be maintained as an overarching 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 from climatology, ecology, biology, pathology, *etc.* Fourth, industrial and government stake-holders, including the public, must be identified in order to determine new data sets and materials to which to transfer and to apply the preceding methodologies and knowledge frameworks for the broadest impact of the ACM framework on industries of the future.[56] Regular workshops and reports, first launched by the National Science Foundation in Washington, DC, in April of 2019 and held again later in May 2020, and embodied in this Perspective, are several such examples. Broader impacts on the public, including technological interests and ethical concerns, must be identified and communicated early and clearly throughout the scientific discovery process: next-generation sensing for safe and reliable autonomous vehicles, heart and brain monitors for health and disease monitoring and treatment, and ecological preservation to avert impacts of global warming are examples of domains that stand to benefit by transformations in our ability to compute autonomously in diverse materials and environments, akin to the human brain.

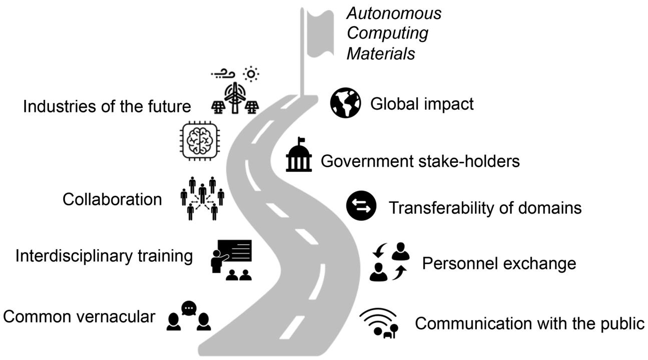

Figure 2. Proposed collaborative, interdisciplinary strategy to enable the integration of neuroscience, data science, and materials science to discover and to disseminate new modes of data storage, sensing, and computing with autonomous computing materials. Activities and initiatives shown are meant to be pursued concurrently, rather than chronologically.

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 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 enable us to move toward the biomimetic architecture of the brain. Placing these pieces together with the tools of the data science revolution suggests the feasibility of designing novel materials that can carry out 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 through voltages and chemical neurotransmitters. Such autonomous computing holds promise for both the construction of large-scale high-performance computing devices and small portable devices capable of being integrated into fabrics and the environment. Here, we have provided a preview of the underlying science of these devices and the possible technologies that it will enable. In a timely quote by R.S. Williams from Hewlett-Packard Laboratories 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 respond to this challenge.

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All authors contributed equally to this article.

Notes

The authors declare no competing financial interest.

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