

Materials for Quantum Technology



ROADMAP

2023 roadmap for materials for quantum technologies

OPEN ACCESS

RECEIVED
28 January 2022

REVISED
20 September 2022

ACCEPTED FOR PUBLICATION
17 November 2022

PUBLISHED
25 January 2023

Original content from this work may be used under the terms of the [Creative Commons Attribution 4.0 licence](#).

Any further distribution of this work must maintain attribution to the author(s) and the title of the work, journal citation and DOI.



Christoph Becher^{1,*} , Weibo Gao^{13,14,*} , Swastik Kar^{3,*} , Christian D Marciniak⁴ , Thomas Monz^{4,5} , John G Bartholomew⁶ , Philippe Goldner⁷ , Huanqian Loh⁸ , Elizabeth Marcellina⁹ , Kuan Eng Johnson Goh^{8,10} , Teck Seng Koh⁹ , Bent Weber⁹ , Zhao Mu² , Jeng-Yuan Tsai³ , Qimin Yan³ , Tobias Huber-Loyola¹¹ , Sven Höfling¹¹ , Samuel Gyger¹² , Stephan Steinhauer¹² and Val Zwiller¹²

¹ Fachrichtung Physik, Universität des Saarlandes, Campus E2.6, Saarbrücken 66123, Germany

² Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore

³ Department of Physics, Northeastern University, Boston, MA 02115, United States of America

⁴ Universität Innsbruck, Institut für Experimentalphysik, 6020 Innsbruck, Austria

⁵ AQT, Innsbruck 6020, Austria

⁶ Centre for Engineered Quantum Systems, School of Physics & Sydney Nanoscience Institute, The University of Sydney, Sydney, Australia

⁷ Chimie ParisTech, CNRS, PSL University, Institut de Recherche de Chimie Paris, Paris, France

⁸ National University of Singapore, Singapore

⁹ Nanyang Technological University, Singapore

¹⁰ Agency for Science, Technology, and Research (A*STAR), Singapore

¹¹ Technische Physik, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

¹² KTH Royal Institute of Technology, Stockholm 106 91, Sweden

¹³ The Photonics Institute and Centre for Disruptive Photonic Technologies, Nanyang Technological University, Singapore 637371, Singapore

¹⁴ Centre for Quantum Technologies, National University of Singapore, Singapore

* Authors to whom any correspondence should be addressed.

E-mail: christoph.becher@physik.uni-saarland.de, wbgao@ntu.edu.sg and s.kar@northeastern.edu

Keywords: quantum, materials, quantum technology, quantum information science

Abstract

Quantum technologies are poised to move the foundational principles of quantum physics to the forefront of applications. This roadmap identifies some of the key challenges and provides insights on material innovations underlying a range of exciting quantum technology frontiers. Over the past decades, hardware platforms enabling different quantum technologies have reached varying levels of maturity. This has allowed for first proof-of-principle demonstrations of quantum supremacy, for example quantum computers surpassing their classical counterparts, quantum communication with reliable security guaranteed by laws of quantum mechanics, and quantum sensors uniting the advantages of high sensitivity, high spatial resolution, and small footprints. In all cases, however, advancing these technologies to the next level of applications in relevant environments requires further development and innovations in the underlying materials. From a wealth of hardware platforms, we select representative and promising material systems in currently investigated quantum technologies. These include both the inherent quantum bit systems and materials playing supportive or enabling roles, and cover trapped ions, neutral atom arrays, rare earth ion systems, donors in silicon, color centers and defects in wide-band gap materials, two-dimensional materials and superconducting materials for single-photon detectors. Advancing these materials frontiers will require innovations from a diverse community of scientific expertise, and hence this roadmap will be of interest to a broad spectrum of disciplines.

Contents

1. Introduction	3
2. Trapped-ion quantum technologies	6
3. Rare-earth ions for optical quantum technologies	9
4. Atom arrays for quantum simulation	13
5. Donor-based silicon quantum technologies	17
6. Color centers in wide-bandgap materials for quantum applications	21
7. 2D materials for quantum technologies	25
8. III–V optical active semiconductor QDs for quantum technologies	28
9. Superconducting materials for single photon detectors	30
Data availability statement	32
References	32

1. Introduction

Christoph Becher¹, Weibo Gao^{2,4} and Swastik Kar³

¹ Fachrichtung Physik, Universität des Saarlandes, Campus E2.6, Saarbrücken 66123, Germany

² Centre for Quantum Technologies, National University of Singapore, Singapore

³ Department of Physics, Northeastern University, Boston, MA 02115, United States of America

⁴ The Photonics Institute and Centre for Disruptive Photonic Technologies, Nanyang Technological University, Singapore 637371, Singapore

The twenty-first century is the age of quantum technologies, where the foundational principles of quantum physics such as coherence, superposition and entanglement of quantum states are tapped—often in devices as small as single defects and impurities—for trillion-times faster computations, beyond-classical ultrasensitive sensors, physically secure communications, and much more. While the scientific community and industry have made giant strides in some of these technologies, others are at earlier stages of technological readiness. In all cases, advancement of these technologies to the next level will benefit substantially from materials advances and innovations. The goal of this roadmap is to identify some of these key challenges and provide insights on material innovations required to ‘move the needle’ further for a range of exciting quantum technology frontiers. In doing so, while we touch upon some relevant recent advances, this roadmap is not meant to be a review article or a progress report.

Innovations in quantum technologies traditionally (and largely) reside within the domain of physics and electrical engineering, with primary applications in the field of information science and technology. However, we hope that this roadmap will appeal to a wider range of disciplines that encompass advancing materials science and engineering. Upstream in the innovation pipeline, materials scientists will play important roles in developing new or upgrading current methods for precision synthesis, scalable yield, and stable performance. At various levels of technological development, new tools and methods are required for difficult-to-diagnose materials problems and to reveal new opportunities. New methods and protocols might be needed to reduce noise and improve reliability. Developed technologies, while addressing traditional information science and related applications, could also potentially advance chemical, biological and other physical sciences. At each point, new advances in data mining and machine learning approaches will benefit materials science. Overall, this roadmap provides a guideline to multidisciplinary research groups and diverse funding agencies as to the crucial advances needed, where the specific needs are, and how best to approach and address these needs.

In our roadmap we cover the most promising material systems in recent applications of quantum technologies. These not only include ‘obvious’ material classes like silicon, diamond, rare earth-doped crystals, semiconductor quantum dots, and two-dimensional (2D)-materials, but also fields where materials play a supportive or enabling role, as for trapped ions or neutral atom arrays.

Trapped ions form one of the most promising avenues for advancing quantum computing and other technologies. The universal control of trapped ion qubits remains an exciting area of growth and requires precision control of trapping structures and laser/microwave/matter interactions. In a typical device, qubits based on trapped ions are mechanically well isolated. However, their control is intimately related to the structural and functional landscape of its nearby materials. Gate architectures and their materials form a key element of these technologies and provide areas of innovation. Quantum enhanced sensing is another attractive area of application using these architectures, giving rise to the possibility of measuring minuscule electrical and magnetic fields in materials and devices. Scaling up the trapped ion platform to a larger number of qubits takes advantage of new technologies and architectures like surface traps, trapping at cryogenic temperatures and charge coupled device (CCD) architecture, giving rise to the development of compact high fidelity quantum CCD simulators [1]. The range of possible applications will greatly benefit from identifying key materials and engineering challenges in these devices and technologies.

Rare earth ions in suitable crystalline matrices can also be exploited for various quantum technologies. Their unusual property that optically active $4f$ transition levels in these systems are naturally protected by the filled outer shells enables them to be naturally isolated from surrounding perturbations. These systems often possess ultranarrow optical transition linewidths and long coherence times at low temperatures. Transitions in Er-based systems arise at or near the 1500 nm telecom wavelength, making them attractive for coupling with existing technologies. However, their weak emission rate means additional signal enhancement architectures are needed for single ion readouts [2]. This system holds the promise to address niche problems in computation, memory, and metrology; challenges in controlled manufacturing of rare earth qubits beyond the laboratory will need to be addressed.

With the development of optical tweezing technology, the possibility of harnessing ultracold arrays of neutral atoms has also become a similarly exciting venue for developing quantum logic devices [3]. Advances

in this field have largely leveraged the exquisite control of individual neutral atoms or molecules made possible by tightly focused lasers. With the tremendous progress in micro/nanofabrication techniques over the past decades, this process can be replicated to enable trapping of arrays of individual atoms. These atom arrays form a powerful method for simulating emergent phenomena and hold the potential for a variety of quantum technologies.

By virtue of its very advanced engineering history, doped silicon will always remain an attractive material for quantum technologies from a commercial scaling and high-quality material availability point of view. Isolated P donors in isotopically pure ^{28}Si form attractive qubits with spin coherence lasting hundreds of microseconds. Both electron and nuclear spins associated with the dopant P could be candidates for quantum computing applications. Low-frequency noise in these systems appear to be extremely low, making them highly attractive for electronically accessible quantum information science. Recent advances in scanning tunneling microscopy (STM)-assisted hydrogen depassivation lithography [4] has enabled engineering of P atoms in the Si matrix with atomic precision—making scalable engineering of Si:P qubits a viable scalable qubit in the near future. Developing these and other high-precision engineering techniques would be a matter of great interest to the quantum technology community.

Single defect centers in wide-band gap semiconductors, such as diamond or SiC to name the most prominent ones, have received particular attention for quantum technology applications due to their often unique combination of long spin coherence times, homogeneous and narrow optical transitions, and bright single photon emission. The solid-state nature of their hosts enables their integration into micro- and nanometer-sized structures for enhanced coupling to optical fields or sensing applications. The benefit of established solid-state technologies lies in the option of miniaturization and scalability, and hence attractiveness for applications in fields where size, weight and power consumption are relevant. On the other hand, undesired accompanying material defects, local strain fields, surface damages or imperfect reconstructions, nuclear spin baths and many more pose limitations on the advantageous properties of the defect centers and call for improvement in material fabrication and processing. Within this field, the benchmark is set by the negatively charged nitrogen–vacancy (NV^-) center in diamond: NV-based quantum sensors defined new application frontiers including biology and condensed matter physics and pioneering proof-of-principle demonstration of quantum network and memory-based communication nodes have been reported. Along with these advancements in diamond color centers, both the optical and spin properties of silicon carbide emitters are optimized by exploiting the matured nanofabrication techniques, optimized quantum control protocol, and material engineering [5].

Single optically active semiconductor quantum dots in recent years have assumed a leading position as sources of single and entangled photons [6] featuring large photon emission efficiencies, high single photon purity and robust entanglement fidelity. These advantageous properties make them interesting candidates for applications in quantum key distribution and quantum networks. The process of quantum dot production, *e.g.*, self-assembled growth, however, leads to large fluctuations of their optical properties, *i.e.*, including variations in emission wavelength or level splitting. Furthermore, the solid state environment gives rise to charge-induced fluctuations of the optical emission (spectral diffusion) and magnetic-noise induced short spin coherence times. In this system, the material challenges thus include optimized growth techniques, material stability, and reduction of charge noise by depleting carriers in electronic diode structures.

In recent times, spectacular progress in the high-quality synthesis and manipulation of 2D materials, and scalable engineering of defects, impurities and excitonic states, has opened up new pathways for niche applications in quantum technologies [7]. Owing to their 2D nature, qubits hosted are ideally suited as quantum sensors with high spatial resolution and high sensitivity based on the small distance to a sample placed on the material surface. Defects in high-bandgap 2D materials such as hexagonal boron nitride (h-BN) have received significant attention as single photon emitters (SPEs), and there is a concerted effort by many groups to predict other defects and 2D materials which might possess the right combination of transition energies and coherence timescales of other quantum technologies. This material system can be considered a fresh entrant in the quantum information science field, and hence many theoretical predictions exist. It remains to be seen if, beyond methods for developing high-quality 2D materials, engineering specific types of defects can also be sufficiently advanced, so as to meet the needs of various quantum technologies.

Photons are an integral part of a variety of quantum technologies, and single photon detection is crucial for efficient quantum information science. All protocols for quantum communication rely on the faithful detection of single photons; figures of merit are the detection efficiency determining *e.g.* the rate of secret key generation and the signal-to-background/noise ratio determining the communication range and fidelity of quantum state transmission. Only the advent of ultra-low dark count and highly efficient superconducting detectors [8] has made possible demonstrations of quantum key distribution (QKD) over hundreds of kilometers. On a more fundamental side, both properties enable closing loopholes in fundamental tests of quantum mechanics (via Bell-type inequalities) which in turn paves the way for advanced quantum

communication protocols like (measurement-) device-independent QKD. Furthermore, superconducting single photon detectors (SSPDs) are an enabling technology for chip-based photonic quantum computing; here the quantum state of photons has to be determined fast and with high fidelity to allow for feed-forward operations in measurement-based ‘one-way’ quantum computing. The challenges on the material side are the optimization of superconducting properties (still less noise, higher efficiency over broad wavelength ranges, less dead time etc) but also the hybrid integration of SSPDs with materials for photonic integrated circuits (silicon, silica, lithium niobate etc).

As discussed in the preceding sections, quantum technologies strongly relate to material properties of the underlying hardware as well as on materials the properties of which are determined by quantum effects (‘quantum materials’). There exist a number of recent reviews and roadmaps on quantum technologies and related materials. Topics that were sufficiently covered in these reviews (e.g. Giustino *et al* [9] for superconducting and topological materials, as well as semiconductor (Si/Ge) quantum dots (QDs); Burkhard *et al* [10] for semiconductor spin qubits; Adcock *et al* [11] for Si photonics) are intentionally not addressed in detail in our present roadmap. In general, we have attempted to keep our discussions distinct from this and other recent reviews on quantum materials, by keeping our focus on issues that pertain specifically to identified quantum technologies.

The field of quantum technologies has witnessed dramatic evolution over the last two decades. Much of the progress was driven by ingenious ideas, concepts and experimental demonstrations ranging from proof-of-principle to first working prototypes. Further development in the field, independent of the nature of the physical quantum bit systems, will hinge on material development for improving optical and spin coherence, reducing inhomogeneous distributions of physical properties, interfacing quantum bits to the environment and enabling scalability. Progress is required in a large range from material composition and purity, reduction of residual defects, targeted doping and isotopic purification, enhanced surface definition and optimized interfaces at material boundaries to advanced material processing technologies like nanoscale etching, hybrid material integration, robust interfacing to optical and electrical industry-norm connectors, integration with control electronics and packaging into rugged devices. As all individual steps and methods become known or ready to be developed, the path to quantum technologies in field and everyday applications will be widely opened.

Acknowledgment

S K acknowledges funding from the Massachusetts Technology Collaborative award number 22032.

2. Trapped-ion quantum technologies

Christian D Marciniak¹ and Thomas Monz^{1,2}

¹ Universität Innsbruck, Institut für Experimentalphysik, 6020 Innsbruck, Austria

² AQT, Innsbruck 6020, Austria

Status

Trapped charged atoms are an established platform spanning the breadth of quantum technologies from metrology and quantum communication to simulation and quantum computing. A strong case for this platform is the near-perfect environmental decoupling of identical constituents. The following roadmap for trapped-ion platforms focuses on quantum computing but can also be readily adapted toward atomic clocks, quantum networks based on trapped ions, or quantum simulators with charged atoms as many aspects are common between these quantum technology pillars for the trapped-ion platform.

Trapped ions have historically led quantum technologies in terms of performance fidelities, with unmatched state preparation and readout error rates. Trapped ions have the highest measurement and duty cycle among atomic, molecular, optical physics (AMO) platforms, albeit smaller than their solid-state counterparts. Trapped-ion quantum registers are medium-sized compared to leading-edge cold atom arrays or superconducting chips but feature full individual control and full-depth hardware connectivity between constituents [16]. Together with typical ratios of gate to coherence times on par with other leading platforms, this leads to resource-efficient algorithm implementations and leading scores in holistic measures of performance such as quantum volume.

Quantum information is stored either in metastable excited states, in ground states of the respective ion, or in both such as in the *omg* architecture. The metastable state has a lifetime depending on species of seconds to years. Gate operations with error rates much smaller than 1% have been achieved using laser-manipulated qubits with two-qubit gate times of about 100 μs . In ground state qubits transitions are mediated via optical Raman or microwave-driven dipole interactions and have recently demonstrated gate error rates as low as $\sim 10^{-4}$ for single qubits, and $\sim 10^{-3}$ for two-qubit entangling gate operations.

In quantum information processing ions are most often trapped in linear configurations using various electrode designs. Traps can be grouped into three-dimensional (3D) traps (usually based on few elements), and surface traps of many elements that are manufactured using established microfabrication. 3D trap designs are technologically mature, and can stably store linear arrays of 50–100 ions, while high-fidelity, individual control is limited to less than 50. Surface traps (with CCD among them) on the other hand are a new technology that has emerged from technical considerations when scaling the platform to significantly larger qubit numbers. 2D ion structures in 3D traps are pursued primarily for analog quantum simulation, where hundreds of ions with global control are well established.

Trapped-ion systems with their long coherence times and optical manipulation capabilities are naturally suited as network matter nodes, and are being actively integrated with photonic links for quantum communication and distributed computing architectures as shown in figure 1. Modularization, standardization, and pioneering integration of quantum computing hardware into high-performance computing and data centers is still in its infancy.

Current and Future Challenges

The primary challenges for trapped-ion technologies going forward are threefold. First, overcoming the current limitations of gate schemes in intrinsic infidelity, required time, and maintaining the control demonstrated as system size is scaled up. Conceptual problems that have been solved for smaller registers are then becoming major engineering challenges for which the solutions are likely to require novel approaches.

Second, the information density in trap architectures needs to increase. Trapped ions as an architecture are atom-number-limited. It is widely believed that linear chains in a single potential cannot be extended significantly past hundreds of qubits. Segmented and CCD traps allow for much higher qubit density compared to connected linear traps past this point, but suffer from so-called anomalous heating which often presents a dominant error source [17, 18]. All architectures face the constraints that collisions with background gas particles impose. These collisions become increasingly likely as the qubit count grows, reducing both the duty cycle and the longest time a single computation can last. A second effort is moving from implementing qubits to higher-dimensional information, or qudits encoded in a single atom. Similarly, the *omg* architecture [19] allows one to encode several qubits in a single atom and allows for the same atom to take different roles, for example memory, information processor, or sympathetic cooling ion. Utilizing bosonic modes such as the ion motion as information carriers is another path past the number limitation.

Third, one of the major roadblocks is the question of how to scalably provide control as register sizes grow. Light-based, single-site addressing exists, but no approach has emerged as the superior strategy for large-scale deployment. Addressing crosstalk already presents a bottleneck in contemporary quantum computers. Alternatively, segmented or surface traps are frequently manufactured with dedicated interaction zones with either integrated, microfabricated optical or electronic addressing sites, or stationary processing zones through which ions are transported electrostatically. However, these transport-based approaches typically perform slower than macroscopic addressing approaches. For some ion species, integration of optical components is complicated by solarization (photobleaching) caused through short-wavelength radiation. Maintaining low temperatures in the relevant modes is a concern that is becoming increasingly pressing. While laser cooling gives access to near-perfect ground state occupation, this typically relies on techniques that scale poorly in necessary time or implementation effort as the register grows. It is ultimately unlikely that a single processor will provide sufficient qubits for large-scale computation. Enabling distributed computing via, for example, photonic links is currently hindered by low-efficiency or poorly scalable light collection, non-deterministic photon generation, and propagation losses which lead to experiment repetition rates far below individual processor capabilities.

Metastable qubit implementations in some species are restricted to comparatively shallow circuits with current gate schemes despite approaching the natural limit in coherence time. While Raman or microwave qubits can overcome the lifetime restriction, they face mechanical and electrical engineering challenges such as interferometric stability for the former and low-noise, high-slew-rate current driving for the latter. It is clear that the number of gates within the coherence time needs to significantly increase in the coming years, which is likely to require novel approaches to performing gates. More detail on particular challenges can be found in [14, 15].

Advances in Science and Technology to Meet Challenges

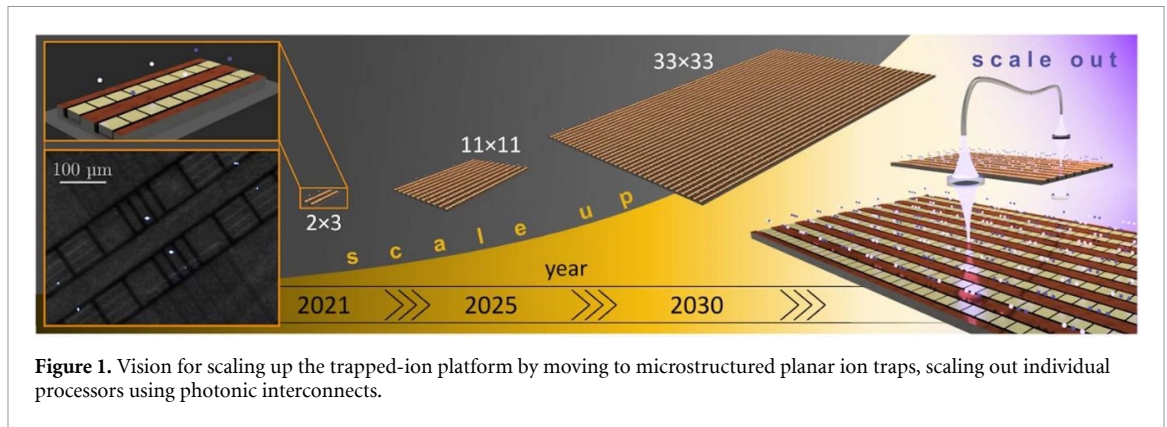
The increasing demands on vacuum quality with growing particle number are likely only to be met by moving from room-temperature setups to cryogenic environments. Management of vibrations through cryo equipment is particularly important for high-fidelity Raman operation. Integrated waveguide and conductor solutions can help to address these new challenges. Scalable distribution of electrical and optical waveguides needs to respect the available space, the low-field-noise environment required, and thus likely require advanced switching and multiplexing capabilities. Maintaining low latencies and minimizing heat conduction is likely to require cryo-compatible, in-vacuum control electronics. As the heat load through active or dissipating elements and higher line density to the outside of the cryostat increases, so does the need for higher cooling power. At the same time, these developments need to be cognizant of the availability of He-3/He-4, whose global and local supply in the face of rising demand may prove a critical limitation rather than just a financial burden.

Cryogenic temperatures may also be required to combat heating in surface trap architectures. While poorly understood, evidence is compelling that this heating originates at the electrode surfaces. Material science and surface treatment technologies need to advance to significantly reduce its magnitude. At the same time, these materials need to be amenable to integration with non-telecom wavelengths, and allow for high slew rate and large arcing voltages to enable faster transport operations of ions. At the same time, microwave-driven gates have the same demands at a very different frequency which further constrains material properties.

Increasing gate speed across all architectures toward nanosecond scales means overcoming the limitations of Coulomb-mediated interactions. Ultrafast Rydberg- or photon-kick-based gates have been realized, but fault-tolerant operation is yet to be demonstrated. Faster gate speeds put even higher demands on shuttling times, as well as photonic links which are unlikely to be met without integrated electronics, and off-the-shelf high-fidelity single photon conversion, collection, and detection.

Seeing that the majority of qubits in ion trap-based quantum computing are currently manipulated by lasers, more reliable, industry-grade lasers face growing demand, including the availability of equipment to stabilize them without intervention for months rather than days. Scalable generation, routing, and distribution of atomic-physics-quality electromagnetic radiation requires advances in massively integrated waveguide technology, amplification, and low-loss, high-extinction switches.

Highly integrated traps will feature many different materials and interfaces between them, many of which will be dielectric. It is as yet unknown how heating caused by these structures will be affected by material choice and ambient temperature. Capacitive and inductive crosstalk from microstructured, integrated circuitry close to the ions needs to be well controlled in order to maintain coherence.



Concluding Remarks

Trapped-ion quantum technologies face many important challenges on their path to large-scale deployment. The prospect of large quantum machines running on naturally identical constituents which offer themselves readily for photonic interconnects in distributed computing or communication is a strong drive in this pursuit. Fortunately, the trapped-ion platform shares many challenges on this trajectory with other leading platforms, from cryostat development to demands on electronics or large-scale integration. Not only continuing to share but increasing synergies between platforms will benefit the quantum technology community as a whole.

Acknowledgements

We gratefully acknowledge funding from the EU H2020-FETFLAG-2018-03 under Grant Agreement No. 820495. We also acknowledge support by the Austrian Science Fund (FWF), through the SFB BeyondC (FWF Project No. F7109), by the Austrian Research Promotion Agency (FFG) contract 872766 and 884471, and the IQI GmbH. This project has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie Grant Agreement No. 840450. T.M. acknowledges funding by the Office of the Director of National Intelligence (ODNI), Intelligence Advanced Research Projects Activity (IARPA), via US ARO Grant No. W911NF-16-1-0070 and W911NF-20-1-0007, and the US Air Force Office of Scientific Research (AFOSR) via IOE Grant No. FA9550-19-1-7044 LASCEM.

All statements of fact, opinions or conclusions contained herein are those of the authors and should not be construed as representing the official views or policies of the funding agencies.

3. Rare-earth ions for optical quantum technologies

John G Bartholomew¹ and Philippe Goldner²

¹ Centre for Engineered Quantum Systems, School of Physics&Sydney Nanoscience Institute, The University of Sydney, Sydney, Australia

² Chimie ParisTech, CNRS, PSL University, Institut de Recherche de Chimie Paris, Paris, France

Status

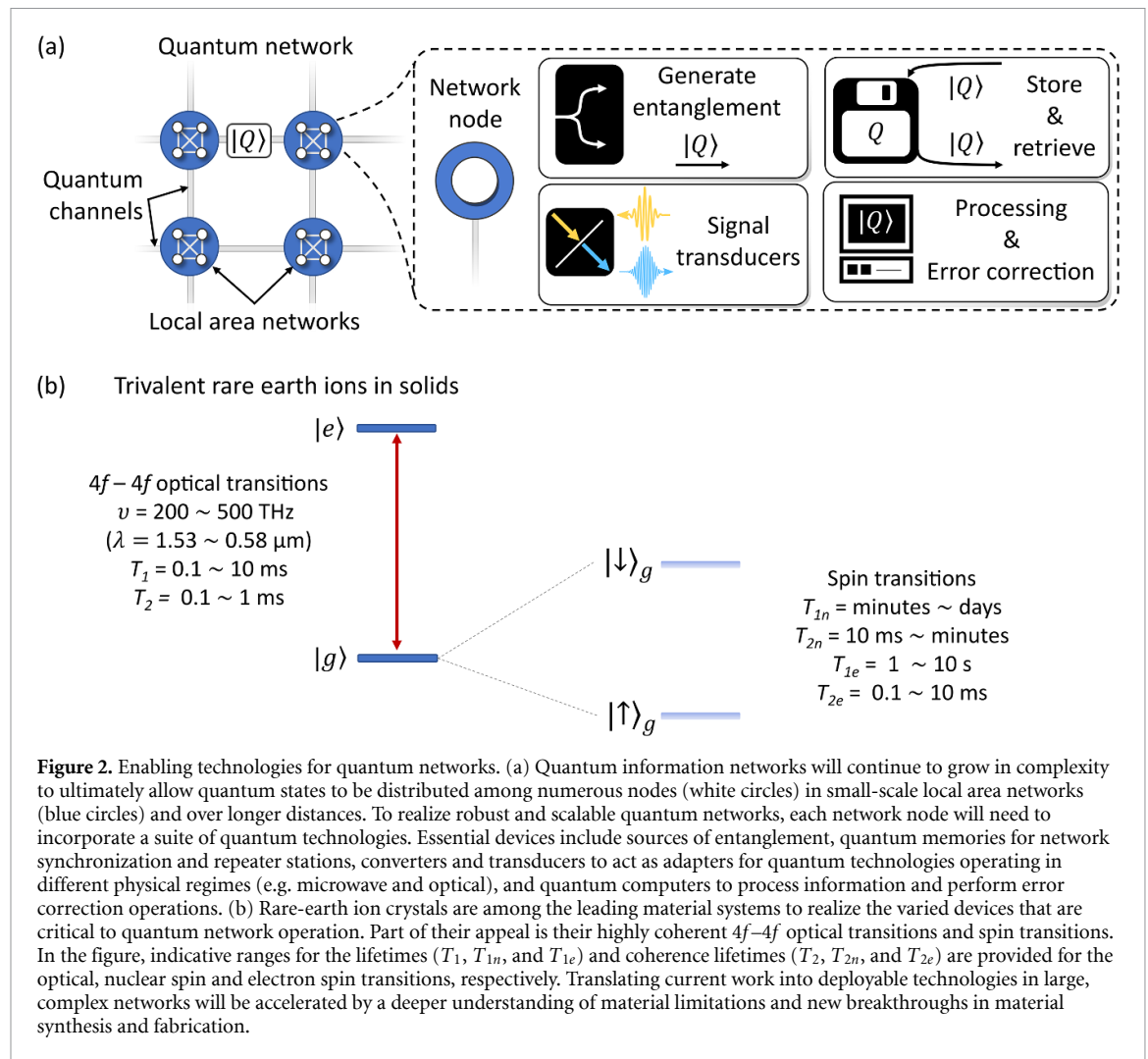
Crystals containing rare-earth ions have a rich history in the field of spectroscopy dating back to the mid-nineteenth century. As purification techniques, laser technology and material science have improved there has been a natural progression from studying the fascinating mechanics of these systems to developing optical technologies. In particular, the pursuit of classical information technologies has generated physical models and spectroscopic techniques that have established a solid foundation for applying these materials to solve challenges in quantum science. The key appeal of rare-earth ions, from single ions [2, 12] right through to large ensembles in stoichiometric crystals [13, 20] is their multiple coherent degrees of freedom. The most prominent is extremely narrow $4f-4f$ optical transition homogeneous linewidths at liquid He temperatures, as narrow as 73 Hz (or coherence lifetime $T_2 = 4.4$ ms) in $\text{Er}^{3+}:\text{Y}_2\text{SiO}_5$ single crystals [21]. Such low broadening is possible because the $4f$ electrons are shielded from lattice perturbations by closed outer electron shells. In addition, rare-earth ions can also possess highly coherent electron or nuclear spin degrees of freedom that can be optically controlled. At suitable temperatures and magnetic fields, these spins can show coherence lifetimes T_2 well into the tens of milliseconds range, and up to 6 h for the nuclear spins of $\text{Eu}^{3+}:\text{Y}_2\text{SiO}_5$ [22, 23].

These distinct properties led to the early recognition that rare-earth crystals are extremely well suited to quantum memory and processor technologies [22]. Rare-earth ion technology is a leading quantum memory platform with demonstrations of long storage times using spin degrees of freedom, high retrieval efficiency, multimode storage, quantum memory entanglement and light matter teleportation [24, 25]. In essence, flying photonic qubits, for example single photons, are mapped to rare-earth optical quantum states through absorption. The quantum information storage time is then governed by the rare-earth optical T_2 . The photonic qubit can then be retrieved on demand using a variety of schemes [22]. Transferring the optical coherence to the spin degree of freedom prior to retrieval allows for large extension of the storage time, a technique known as spin-wave storage. Developed in parallel with quantum memory technology were initial proposals and experiments for quantum processing based on ensembles of ions [13]. Recently, advances in single ion detection, especially by coupling to micro- or nano-optical resonators, have led to single spin coherent control and single shot readout, opening another way to optically addressable spin qubits [2, 12]. The combination of quantum memories and qubit processing nodes could allow quantum information to be distributed over large distances through optical quantum repeater links. Rare-earth ion technology provides the basis for generating, storing, processing and converting information at the quantum level (see figure 2), which are critical elements required for an integrated quantum network.

An additional strength of the rare-earth ion platform is its versatility. For example, the $4f-4f$ optical transitions in different rare-earth ions cover a broad spectral range, from infrared to ultraviolet, including the important telecom wavelength at $1.55 \mu\text{m}$ found in Er. These ions can be introduced in various insulating crystals, glasses, and semiconductors either during synthesis or by implantation. This provides access to a very large number of host-ion combinations, which can be leveraged not only as bulk materials but also as micro- or nano-structured materials such as nanoparticles or thin films [26]. Although many of the achievements place the rare-earth ion system among the best systems for quantum information applications, these successes are still limited by material challenges. Therefore, the removal of these restricting factors is truly an exciting prospect.

Current and Future Challenges

A first important research topic is to identify and grow improved bulk crystals. For example, many currently used crystals limit quantum device performance through the presence of abundant nuclear spins. In yttrium-based hosts, such as Y_2SiO_5 , ^{89}Y has a $1/2$ nuclear spin with 100% abundance and the magnetic noise caused by this spin bath can ultimately limit coherence lifetimes, especially for electron spins. This problem is even worse in hosts like YVO_4 or LiNbO_3 , which contain nuclear spins with large magnetic moments. In addition, nominally undoped Y-based crystals contain unavoidable rare-earth ion impurities at a 0.01–1 parts per million level, which is detrimental to single ion qubit devices. Several crystals with heavily reduced nuclear spin concentration and ultra-low rare-earth background impurities show promising properties such as Er^{3+} -doped CaWO_4 , Si, or TiO_2 [27]. A challenge, however, is that these materials do not

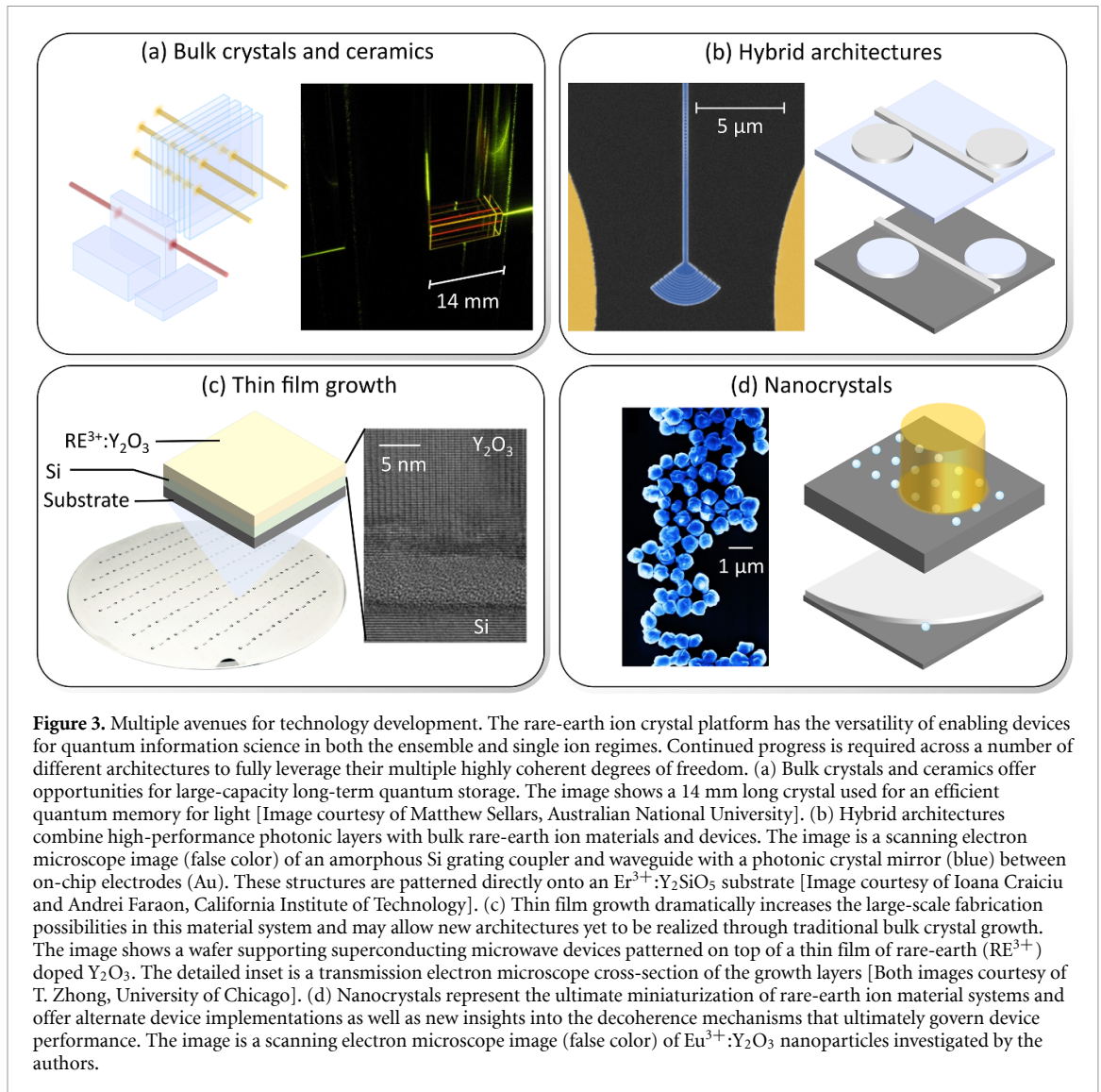


have substitutional sites with well-matched size and charge for trivalent rare-earth ions, which may lead to additional defects and strain and in turn potential sources of decoherence.

Integrating rare-earth ions with photonic or microwave resonators presents another important materials challenge (figure 3). Such coupling is essential in ensemble and single ion devices for high-efficiency and high-fidelity operations. For example, ion–resonator coupling is critical to overcome the weak rare-earth ion optical oscillator strength ($<10^{-6}$) to achieve high-efficiency single ion qubit detection and control. An ideal host would combine low optical or microwave waveguide and cavity loss with optimized rare-earth ion properties like narrow homogeneous and inhomogeneous linewidths. This can be at least partly obtained in crystals well developed for integrated optics like LiNbO_3 or Si [28–31] where rare-earth ions can be introduced by doping or implantation. Structures can also be fabricated directly into crystals well suited to rare-earth ions like Y_2SiO_5 [2]. Hybrid designs with surface resonators or open cavities are also available and could provide optimized optical and rare-earth properties at the same time [12, 32].

Attempting to increase the coherent ion–resonator coupling rate in integrated systems by minimizing the resonator mode volume increases the ions’ proximity to interfaces and the likelihood of increased decoherence. This key trade-off is common to all architectures for miniaturized devices including resonators that are milled, etched or deposited onto the surfaces of bulk crystals. Efforts to overcome this trade-off include mitigating near-surface electric field perturbations by using rare-earth ions in non-polar site symmetries in which permanent electric dipole moments vanish for all electronic levels. This reduces electric field broadening at the expense of one avenue for two-qubit gate operations and the ability to frequency tune using electric fields.

Another option is to improve material synthesis, fabrication, and post-processing, concentrating specifically on the crystal interfaces. The synthesis of rare-earth ion nanoparticles or thin films offers a way to study the optimization of these material improvements as well as appealing alternate architectures for devices, such as coupling to open micro-cavities. However, rare-earth ions in nanomaterials generally show



degraded properties, most notably optical T_2 , compared to single crystals. Efforts to circumvent these problems are currently focusing on improving synthesis and post-processing of nanomaterials [26].

High-resolution and controlled spatial localization could be achieved with implantation in hosts with low background rare-earth impurities. As rare-earth ions are heavy elements it is expected that significant damage occurs during implantation. It has been shown that annealing can at least partially cure these defects, leading to relatively narrow lines [28–31]. It is, however, unclear to what extent implanted ion properties can approach those of as-grown materials.

Advances in Science and Technology to Meet Challenges

A fundamental requirement of efficiently optimizing materials is to be able to link structural information with relevant properties. In the case of rare-earth ion coherence lifetimes, accurate models have been developed for bulk crystals in regimes where the rare-earth or host electron spins are the main source of dephasing. The roles of defects, impurities and residual disorder are less clear and often difficult to relate to structural analyses but there is still likely to be a limit at low doping concentration or in spin-free hosts. For ions close to the surface, charge noise is a likely cause of excess dephasing and spectral diffusion compared to the bulk ions but direct experimental evidence is yet to be demonstrated. In the same way, the relatively short optical T_2 in nanoparticles and thin films are likely to be due to charge noise at the surface and also within the lattice as a result of a higher concentration of defects such as oxygen vacancies. A deep understanding of these different results is likely to emerge from comprehensive studies that involve a range of materials and techniques like XRD, electron microscopy, chemical analysis, electron paramagnetic resonance, luminescence of defects, and their correlation with rare-earth spectroscopy. From this understanding, optimized synthesis

of materials and post-processing can be designed starting from the broad range of techniques available for bulk and nanoscale materials.

In the case of implanted samples or nanomaterials, low rare-earth concentration or material volume results in low signals, which makes rapid feedback toward material development more difficult. Here, experimental throughput could benefit from progress in low-temperature microscopy and high-sensitivity superconducting nanowire detectors. An even more difficult investigation is single ion spectroscopy, which currently requires material-specific nanofabrication techniques. The development of easy to operate, high-throughput experimental techniques such as open optical micro-cavities or samples integrated with superconducting detectors could solve this issue for nanomaterials and bulk samples thinned down to micrometer-thin membranes.

Hybrid approaches to rare-earth integration into photonic structures could be improved in several ways. As stated above, material coherence properties at the nanoscale should first be improved. Second, high-quality light waveguides and resonators adapted to rare-earth wavelengths in the near infrared or visible regions would enable exploiting new rare-earth transitions beyond the silicon compatible Er^{3+} telecom wavelength. Another technology of potentially broad applicability is laser written photonic structures, which seem to affect rare-earth coherence only to a small extent [33].

Concluding Remarks

Rare-earth ions in crystalline hosts are promising systems for optical quantum technologies, showing optical and spin transitions with some of the longest coherence lifetimes demonstrated in the solid state. Further use of these exceptional properties toward integrated quantum photonic architectures by leveraging the ensemble and single ion regimes requires significant improvement in material engineering, as coherence lifetimes can be reduced by order of magnitudes compared to bulk materials at the nanoscale. Such developments should be guided by a comprehensive understanding of the dephasing mechanisms involved. Otherwise, novel high-quality photonic structures that can couple with rare-earth crystals are needed. As the latter can be obtained in different forms (bulk, nanoparticles, thin films) or and operate at different wavelengths, a variety of successful combinations can be expected for different applications. In this respect, the search for new material platforms, such as rare-earth molecular crystals [34], or through atom-scale precision site engineering, could enormously broaden the scope of possible systems.

Acknowledgements

The authors would like to acknowledge the support of the Australian Research Council Centre of Excellence for Engineered Quantum Systems (EQUS, CE170100009), and of the European Union's Horizon 2020 research and innovation programme under Grant Agreement No. 820391 (Square).

4. Atom arrays for quantum simulation

Huanqian Loh

National University of Singapore, Singapore

Status

Ultracold atoms individually trapped in arrays of optical tweezers have rapidly emerged as a powerful platform for quantum simulation of many-body physics, which underlie the quest for a basic understanding of advanced materials. These atom arrays offer scalable single-qubit control over both the internal spin state and the external motional quantum state. They can also be generated in arbitrary geometries in one, two, or three dimensions with programmable doping and tunable interactions. These combined features of scalability and programmability render them highly attractive for analog quantum simulation, where the atom arrays are set up to obey the same Hamiltonians used to model materials.

To generate the atom arrays, atoms are first stochastically loaded from optical molasses into an array of optical traps. These optical traps are formed by sending the trapping laser through either a spatial-light modulator or acousto-optic deflectors, and then focused onto the atoms using a high-numerical-aperture microscope objective. At this stage, the filling fraction is usually limited by collisional blockade to be about 50%–60%. The filled trap positions are imaged using atom fluorescence and then fed back in real time to acousto-optic deflectors that generate one or more moving tweezers to rearrange the atoms by dragging them to target sites (figure 4). Such dynamic reconfiguration has enabled the creation of defect-free atom arrays in user-defined geometries ranging from honeycomb lattices [35] to 3D fullerene-like arrays [36].

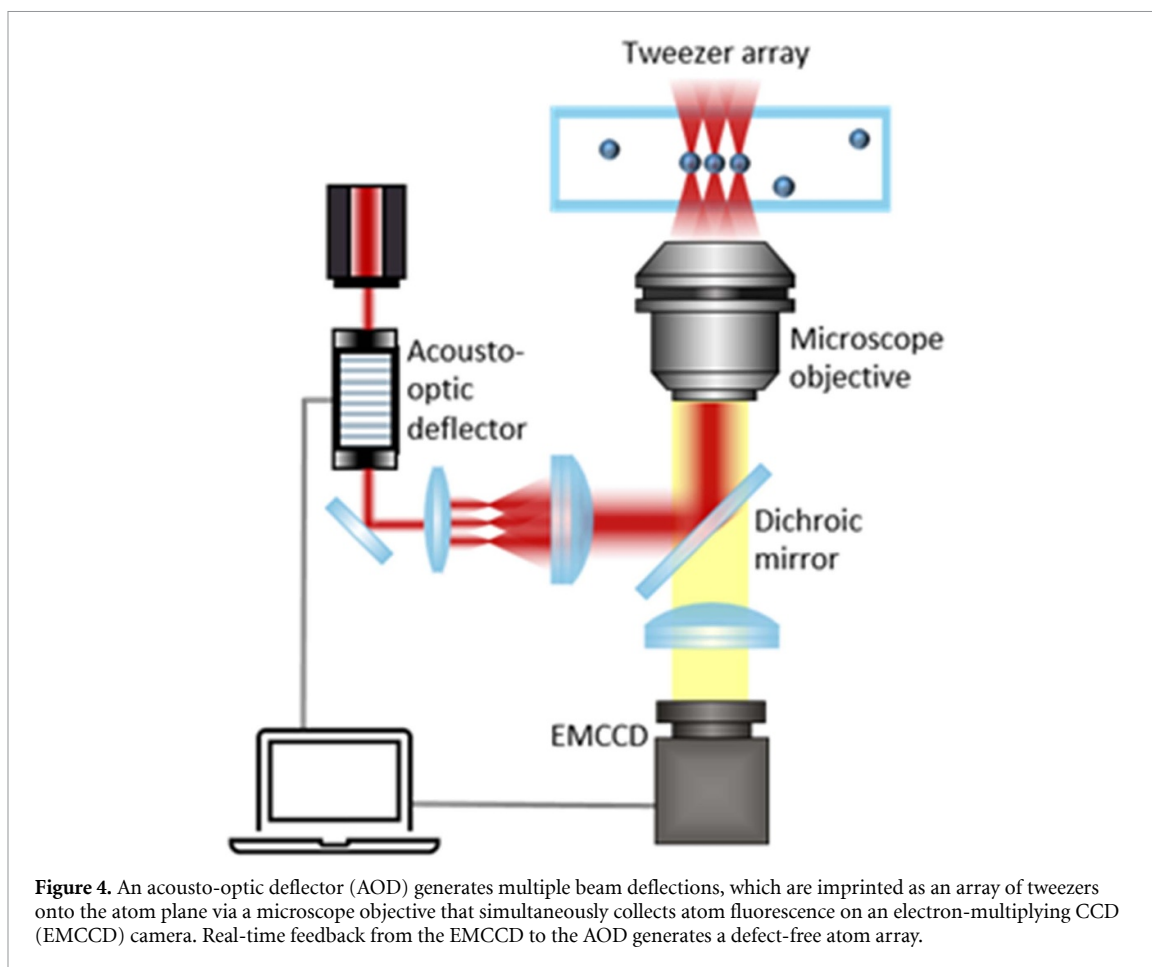
To map the defect-free array onto a given model Hamiltonian, tunable interactions need to be induced between the singly trapped atoms. Exciting atoms to Rydberg states (principal quantum number ~ 60 – 80) has emerged as a natural route for inducing tunable interactions in atom arrays, as the Rydberg interactions V/\hbar can remain on the scale of 1–10 MHz for typical tweezer spacings of several micrometers. Such strong Rydberg interactions can be induced via one of the following ways:

- Coherently driving the array of atoms between the ground state $|g\rangle$ and a Rydberg state $|r\rangle$ with a laser resonant to the ground–Rydberg transition frequency for a single atom;
- Transferring the atoms to the Rydberg state using stimulated Raman adiabatic passage and subsequently driving the atoms between two Rydberg states $|r_1\rangle, |r_2\rangle$.

The first method induces van der Waals interactions between atoms in the same Rydberg state. These interactions scale as C_6/r^6 , where r is the distance between atoms. When an atom array is uniformly illuminated by a resonant Rydberg laser of Rabi frequency Ω , Rydberg blockade occurs, where atoms within a given blockade radius $R_b = (C_6/(\hbar\Omega))^{1/6}$ of a Rydberg atom are prevented from being excited to the Rydberg state as their transition frequencies are shifted off resonance. Mapping the ground state and Rydberg state onto $|\downarrow\rangle$ and $|\uparrow\rangle$ respectively allows one to implement an Ising-type Hamiltonian with the atom array [37]. By ramping the detuning adiabatically across the Rydberg transition resonance, crystalline phases of Z_n symmetry have been observed, where n refers to the number of neighboring blockaded atoms and is controlled by varying the array spacing relative to the blockade radius [38]. Beyond phase transitions, the atom array is also well suited to studies of spin dynamics, as the quantum states of individual atoms in the array can be detected directly through atom fluorescence. For instance, dynamical signatures of quantum many-body scars have been observed by bringing the atom array to a rapid quench and seeing the atoms exhibit long-lived coherent oscillations between the states $|\downarrow\uparrow\downarrow\uparrow\dots\rangle$ and $|\uparrow\downarrow\uparrow\downarrow\dots\rangle$ [38].

The second method of Rydberg excitation induces dipolar interactions between atoms in the two different Rydberg states. These dipolar interactions are both long-range (scaling as C_3/r^3) and anisotropic. Coupling between the two Rydberg states is typically achieved using a microwave field. Spin models like the XY and XXZ spin Hamiltonians can be engineered by mapping the two Rydberg states onto $|\downarrow\rangle$ and $|\uparrow\rangle$ [37]. For instance, the Su–Schrieffer–Heeger model, initially proposed to explain the high conductivity of doped organic polymers, was realized for a one-dimensional atom chain by arranging the Rydberg atoms such that next-nearest-neighbor hoppings are suppressed by the anisotropy of the dipolar interactions, whereas nearest-neighbor couplings alternate between a strong and a weak value (figure 5) [39]. This led to an observation of a symmetry-protected topological phase for interacting bosons [39].

So far, the above methods of inducing tunable interactions have involved direct excitation of atoms to the Rydberg state. While these Rydberg states boast large interaction strengths, their lifetimes are typically limited to hundreds of microseconds. A complementary method of Rydberg excitation involves admixing the ground state with a small component of the Rydberg state by using an off resonant dressing laser, where the Rabi frequency is small compared to its detuning. This Rydberg dressing allows for interactions between two



long-lived spin states in the electronic ground state and offer possibilities for realizing more sophisticated spin Hamiltonians [37].

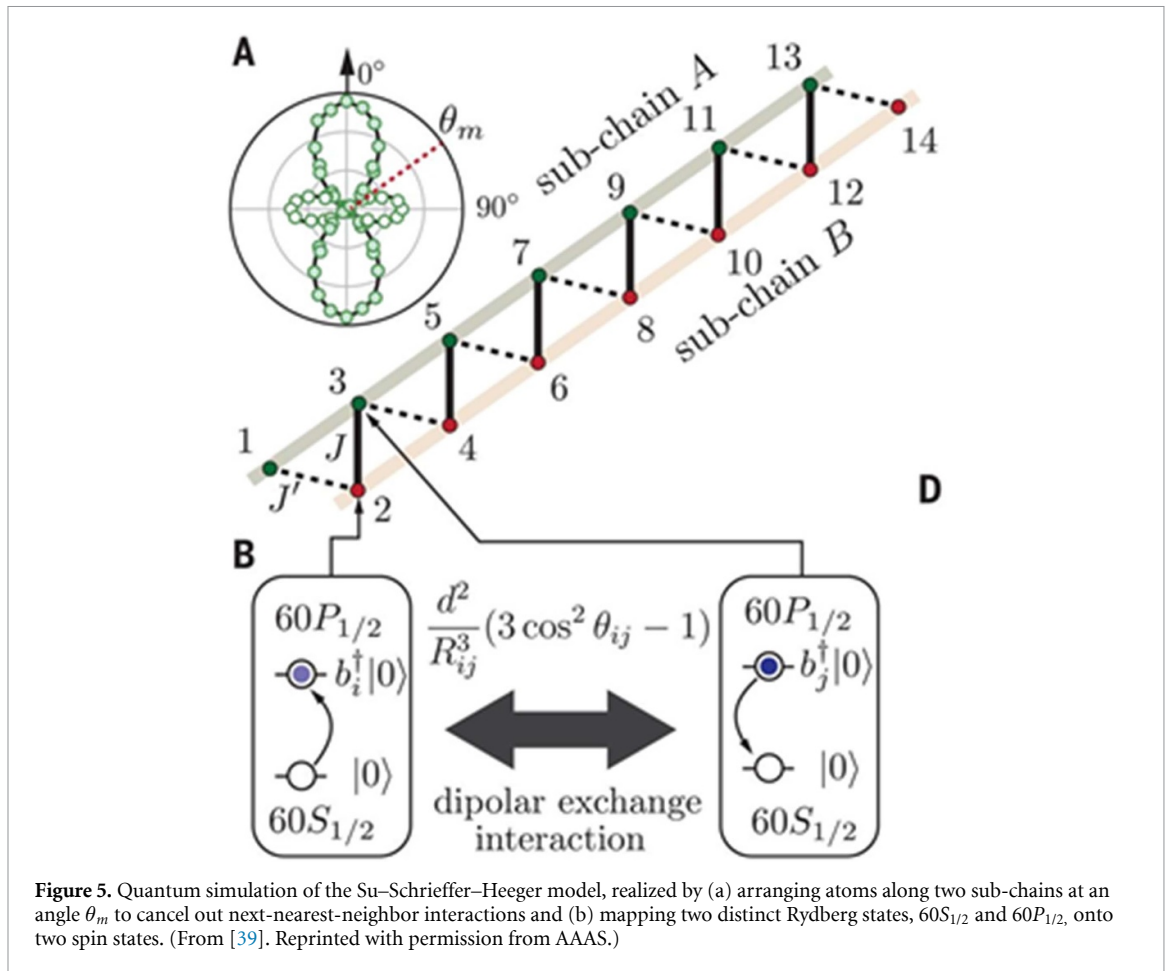
Current and Future Challenges

At present, state-of-the-art quantum simulation experiments with atom arrays involve hundreds of singly trapped atoms [35]. Scaling the size of defect-free arrays up to thousands of atoms remains a significant challenge. The array size is typically limited by tweezer laser power, which is in turn related to a combination of factors like the minimum trap depth required to load and image atoms, atom polarizability at the chosen tweezer wavelength, and availability of high-power lasers. Concurrently, as the array size increases, high-fidelity rearrangement of atoms to generate defect-free arrays becomes more difficult. This is because larger arrays tend to require longer reconfiguration times, yet atoms have a finite trap lifetime due to collisions with the background gas, which causes an exponential decay in the probability of a defect-free target array at the end of rearrangement. Therefore, efficient algorithms that minimize the rearrangement time or techniques that suppress the atom loss mechanisms need to be developed.

Besides scalability, establishing a higher degree of control over the atom-array platform would help push the frontiers of quantum simulation experiments. These include better programmability of interactions, higher-fidelity quantum state preparation and readout, and improved spin coherence. Some of these challenges can be tackled by using different atomic species, while others can be mitigated through technical improvements. For instance, where atom-array experiments involve two-color Rydberg excitations, the ground–Rydberg coherence times are limited by Rydberg excitation laser phase noise, spontaneous emission from the intermediate state, and Doppler effects from finite atom motion. These can be improved respectively by cavity filtering of laser phase noise, using high-power Rydberg lasers to compensate for larger detunings from the intermediate state, and cooling atoms to the motional ground state.

Advances in Science and Technology to Meet Challenges

To scale up atom arrays, efforts have been made to build high-power lasers at conventional tweezer wavelengths by frequency doubling fiber-amplified lasers. A complementary approach is to reduce the trap depth required to load atoms. For alkali atoms, D1 light has been shown to not only cool atoms efficiently



when loading them into shallow traps, but also yield enhanced loading probabilities as high as 90% by driving atoms to a repulsive molecular state [40]. Combining these techniques with D1 magic wavelength tweezers further increases the overall scalability by reducing the trap depth used when imaging atoms [41].

For high-fidelity generation of large defect-free arrays, the atom array can be placed in an ultra-high-vacuum-compatible cryostat to improve the atom lifetime [42]. Such a cryogenic setup can be combined with parallel rearrangement algorithms and the aforementioned methods for enhanced loading probabilities to reduce the rearrangement time. The cryogenic temperatures would also suppress black-body radiation-induced transitions, which can lead to longer Rydberg lifetimes and Rydberg-dressing coherence times.

Beyond the alkali atoms typically used in atom-array experiments, alkaline earth atoms have recently been pursued for their increased coherence and high-fidelity state preparation and readout methods [43]. Multiple atomic species can also be combined in a single setup to explore alternative ways of inducing programmable interactions, such as by combining the pre-cooled atoms to form ultracold dipolar molecules in tweezer arrays [44]. Such singly trapped molecules, whose electric dipole moments can be manipulated in the ground state using both electric fields and microwaves while avoiding chemical reactions with other molecules, offer rich possibilities for realizing exotic Hamiltonians and probing spin dynamics with long coherence times.

Finally, tweezer arrays of atoms can be interfaced with nanophotonic chips made out of SiO_2 and Si_3N_4 for enhanced atom-photon coupling and single-atom control [45]. Further, the mediation of entanglement between two atoms via their strong coupling to SiN photonic crystal cavities has been recently demonstrated, and such entanglement has been verified to be preserved even when atoms are transported away from the near field of the cavity [3]. These results make it promising to use nanophotonic cavities as interconnects between remotely spaced atom arrays. Extending this work to the telecom wavelength range [46, 47] would enable large-scale quantum networks of atom arrays over long distances, thereby elevating the applications of atom arrays beyond quantum simulation to include quantum communication and more general quantum information processing.

Concluding Remarks

Atom arrays are one of the leading platforms for analog quantum simulation of many-body Hamiltonians that serve as simplified models of advanced materials. Further improvements in scalability, control, and programmability will not only push the frontiers of quantum simulation but also make atom arrays highly competitive for quantum information processing in general.

Acknowledgements

We acknowledge support from the National Research Foundation (NRF) Singapore, under its NRF fellowship (NRF-NRFF2018-02) and its QEP2.0 programme (NRF2021-QEP2-02-P09), with further support from the Singapore Ministry of Education Academic Research Fund Tier 1 (A-0004210-00-00).

5. Donor-based silicon quantum technologies

Elizabeth Marcellina¹, Kuan Eng Johnson Goh^{1,2,3}, Teck Seng Koh¹ and Bent Weber¹

¹ Nanyang Technological University, Singapore

² Agency for Science, Technology, and Research (A*STAR), Singapore

³ National University of Singapore, Singapore

Status

Silicon has been the cornerstone of the modern semiconductor industry for over six decades, and, since the seminal proposal by Kane in 1998 [48] (figures 6(a)–(c)), has also become a promising platform for quantum technologies [49, 50]. Owing to impressive advances in semiconductor device physics and engineering, the past two decades have seen key demonstrations of quantum processor prototypes, including those in donor-based architectures [50–58] (figures 6(d)–(i)).

The main motivation for utilizing Si as a host material for quantum technologies is rooted in the weak interactions of electron (or nuclear) spins with the crystalline host. The resulting exceptionally long spin lifetime (T_1) and coherence time (T_2) have earned Si the nickname of a ‘semiconductor vacuum’ [59], a condensed-matter analog of cold-atom or trapped-ion systems hosted in ultra-high-vacuum environments. Indeed, the spin–orbit interaction for electrons is generally considered weak [58], and the already weak contact hyperfine fields in natural Si (^{nat}Si) can be further reduced in isotopically purified ²⁸Si. This is particularly true for donor spin qubits in Si, due to their atomic-scale footprint with a tightly confined, spherically symmetric electron wave function.

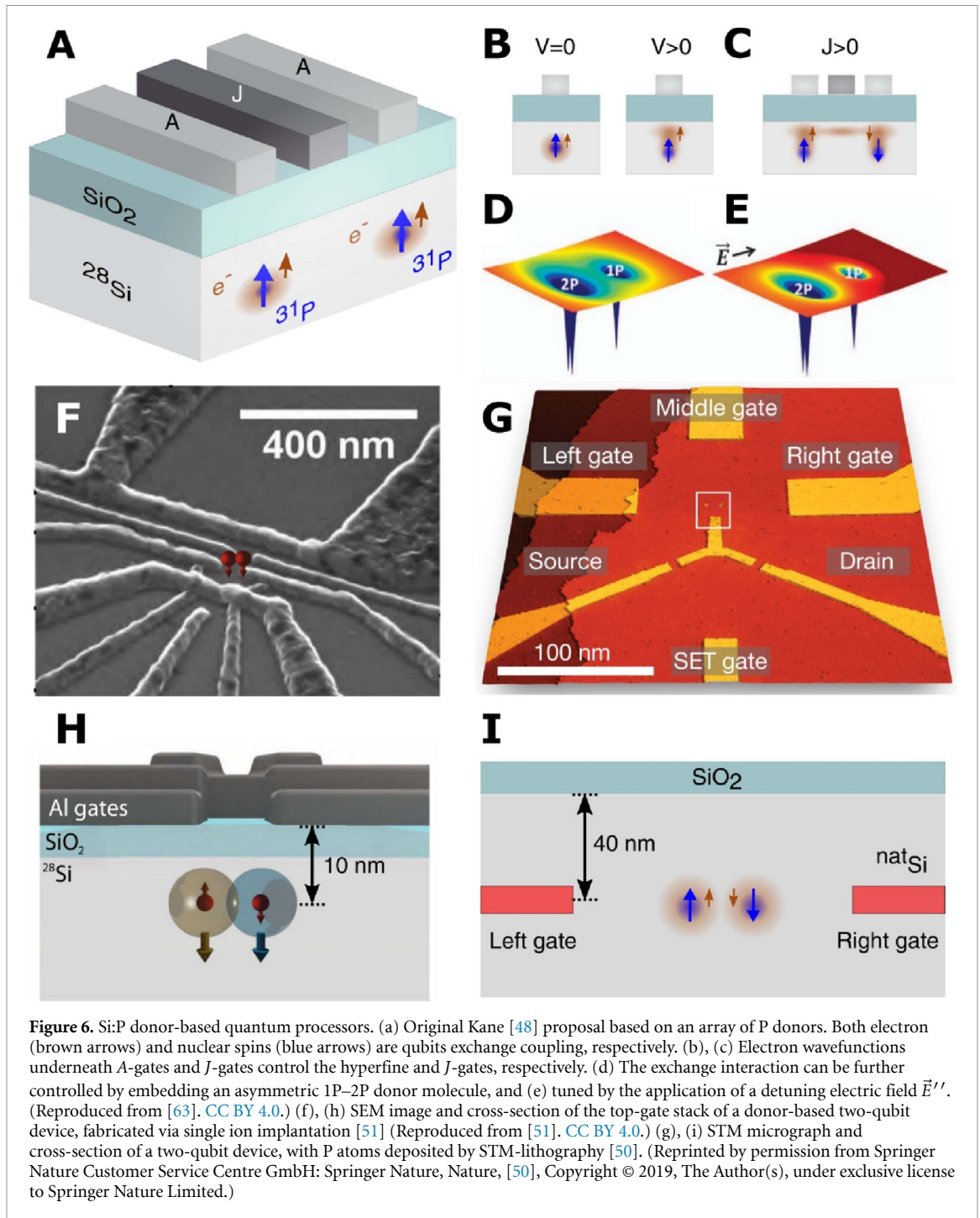
Electron spin resonance (ESR) and nuclear magnetic resonance (NMR) experiments of P (³¹P) bulk donor ensembles date back as early as the 1960s and have shown electron and nuclear spin lifetimes (T_1) of ~ 1 and ~ 10 h [60], respectively, at low temperature, even in ^{nat}Si. Recent measurements of donor ensembles in ²⁸Si have yielded coherence times $T_2 = 10$ s for the electron spin [61]. Dynamically decoupled (DD) coherence times of up to $T_{2DD} = 3$ min have been measured for the nucleus if the donor is in its neutral state [59], or even 180 min if it is ionized [62].

To date, two established technologies exist to realize nanoelectronic devices based on ³¹P donor qubits (figures 6(f)–(i)). Most of the early landmark demonstrations have been achieved by high-throughput ion-implanted metal–oxide–semiconductor compatible device architectures [51–55] (figures 6(f) and (h)). These demonstrations include electron (ESR) and nuclear (NMR) spin resonance of individual qubits, detected in projective single-shot electron spin read-out. More recently, a controlled-rotation (CROT) exchange gate [51], and even an entangled Greenberger–Horne–Zeilinger three-qubit state of two ³¹P donor nuclei coherently coupled to a single electron spin [54], have been demonstrated.

Alternatively, donors can be placed with an accuracy < 1 nm using STM lithography [50, 56–58] (figures 6(g) and (i)). In this approach, the qubits themselves are embedded in the Si single crystal alongside control electrodes, gates, and single electron transistor charge sensors in an all-epitaxial 2D co-planar device architecture [50, 56–58]. The high spatial accuracy in donor placement allows for precision engineering of the strength of the exchange coupling j [56], sensitive to both separation and orientation of a donor–qubit pair in the Si matrix, and has subsequently allowed for the demonstrations of a fast (800 ps), high-fidelity ($\sim 94\%$) exchange gate (figures 6(g) and (i)) [50].

Current and Future Challenges

The single most sizable challenge in donor-based quantum computing is to scale to an appreciable number of qubits. Kane’s original architecture [48] envisioned linear arrays of donor qubits, precisely placed at separations of order 10 nm, dictated by the strength of the exchange interaction. However, for any technique, it remains challenging to embed ³¹P donor atoms into Si accurately, reproducibly, and efficiently toward such scalable arrays. Despite the impressive technological advances in deterministic single ion implantation [64], ion straggle may inherently limit placement accuracy to the ~ 10 nm scale. Implanted P₂ molecules have allowed for demonstrations of two-qubit gates. However, the separation and orientation of the implanted donor molecule cannot usually be controlled, and requires that suitable donor pairs be electrically isolated from large implanted ensembles [51]. At the same time, STM-assisted qubit placement remains limited in throughput and device yield, despite its advantages in accuracy. Moreover, the stochastic nature of the dissociation and incorporation chemistry of the dopant precursor molecule results in donor number fluctuations at qubit incorporation sites, often resulting in the placement of donor clusters rather than individual donor atoms [50, 56]. Alternative precursors for both donors and acceptors have recently been considered, although they have yet to reach comparable maturity with the Si:P-based fabrication. To further

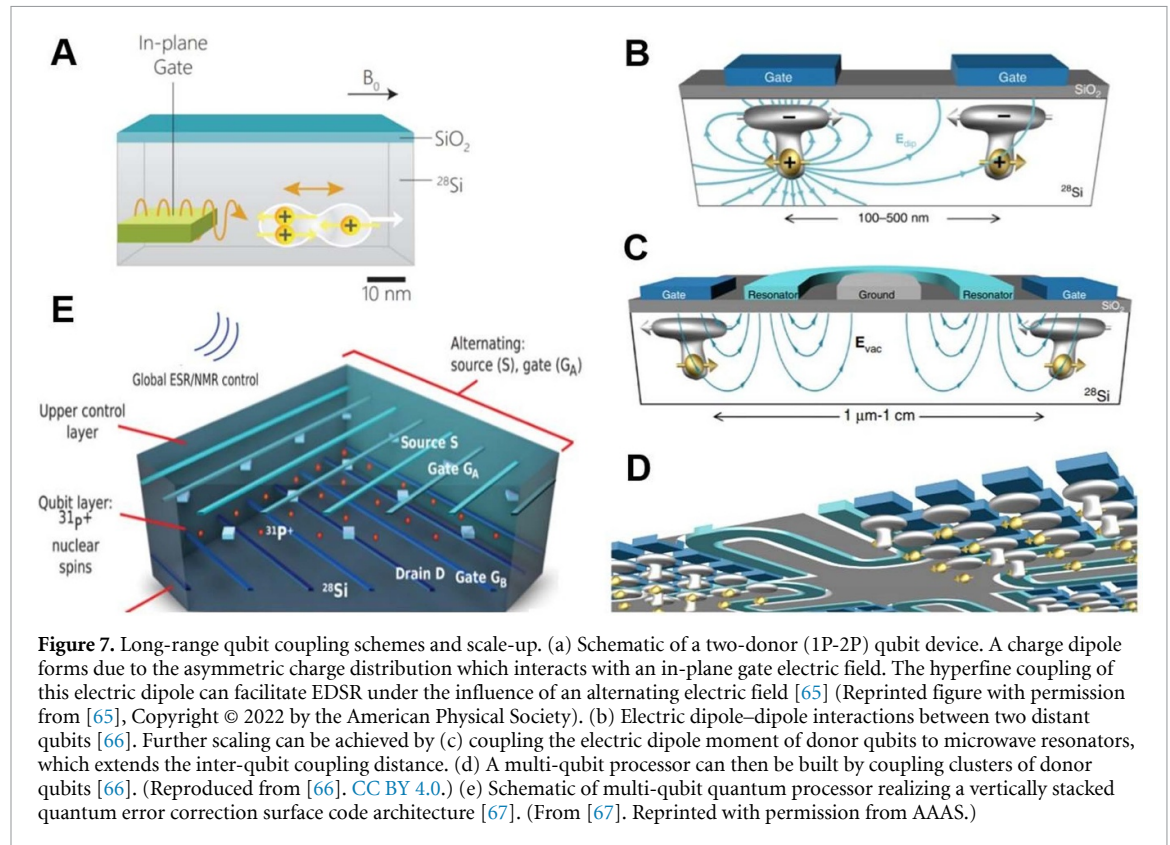


streamline the STM-based fabrication process for fabrication accuracy, efficiency, and reproducibility, process automation including machine learning and artificial intelligence algorithms are likely to be required.

To date, only the ion-implantation approach embeds qubits in isotopically enriched ^{28}Si , which is achieved by high-fluence ^{28}Si ion implantation of the surface layer. Efforts are currently also underway using the STM-assisted approach to achieve epitaxial growth of ^{28}Si in ultra-high vacuum (UHV). However, even in ^{28}Si , electron and nuclear spin coherence times remain significantly shorter (of the order of three magnitudes) in nanoelectronic devices, compared to bulk ensembles (table 1). The stark difference between the individual and bulk coherence times may be attributed to several factors. The presence of surfaces/interfaces in devices may lead to increased charge noise, although placing donor qubits within epitaxial single crystal Si has been shown to reduce noise levels to the shot noise limit [57]. Vicinal donors may allow dipolar flip-flop or possibly even hyperfine coupling of nuclear spins. Lattice strain and electric

Table 1. Summary of spin lifetime (T_1) and coherence time (T_2) measured in donor qubits in nanoelectronic devices (at $T \approx 100$ mK), compared to bulk ensembles. T_{2DD} denotes T_2 enhanced by dynamical decoupling, and T_2^* denotes the Ramsey envelope decay time. The measurement temperatures for the bulk ensembles range between 1 and 3 K.

Qubit type	Bulk ensembles	Nanoelectronic devices
Electron spin (T_1)	>hours [60–62]	>seconds [52]
Electron spin (T_2)	10 s [61] (^{28}Si)	$T_2^* = 268 \mu\text{s}$ ($T_{2DD} = 0.56$ s, ^{28}Si) [55]
Nuclear spin (T_1)	>hours [60, 62]	>days [52]
Nuclear spin (T_2)	$T_2 = 27$ s ($T_{2DD} = 3$ h, ^{28}Si) [62]	$T_2^* = 600$ ms ($T_{2DD} = 35.6$ s, ^{28}Si) [55]



fields are also known to affect spin lifetime and coherence time. The latter in particular, inherent to the gate control of nanoelectronic devices, may lead to an enhancement of the spin–orbit coupling strength [58].

Advances in Science and Technology to Meet Challenges

Even when single donors are placed with atomic level accuracy, for scalable architecture each qubit in the array needs to be addressed individually by local electric and magnetic fields without affecting their neighbors. The combined factors of the atomic-scale footprint of the qubits, the short-range nature of the exchange interaction, and the high gate-density it dictates have all been contributors to the challenge of scaling to more than two donors [50, 51, 54, 65–67]. To overcome scalability constraints, long-range qubit coupling schemes, such as electrical dipolar interactions, are currently being explored [65, 66] (figures 7(a)–(c)). An electric dipole may be formed by biasing a donor wave function close to the ionization point, hence causing a charge separation between the electron and the positive donor nucleus (figure 7(b)) [66]. Alternatively, combining a single donor with a donor molecule creates a finite hyperfine interaction between the two (figure 7(a)) [65] which may then be modulated by time-dependent electric fields to induce electric dipole spin resonance of the electron and nuclear spins. Within this scheme, quantum information may be encoded in the states $\{|\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle\}$, for the single donor [66] and $\{|\uparrow\uparrow\downarrow\downarrow\rangle, |\downarrow\downarrow\uparrow\uparrow\rangle\}$ for the single donor–donor molecule system [65]—forming so-called ‘flip-flop’ qubits. Electric dipole coupling of up to tens of megahertz and distances of hundreds of nanometers have been predicted [65, 66], making this approach a viable long-range alternative in addition to exchange-coupled qubits. Qubit coupling may further be extended to mesoscopic or even macroscopic distances ($\sim 1 \mu\text{m}$ – 1 cm) by coupling of spin qubits to microwave resonators [65, 66] (figure 7(c)), possibly opening avenues for hybrid quantum technologies including both spin qubits and superconducting qubits (figure 7(d)). To further meet the challenges of

scale-up, quantum error correction protocols, including topological surface codes, have been proposed [67]. Surface codes may be implemented within 2D qubit arrays controlled by a 3D network of electrodes (figure 7(e)).

Concluding Remarks

Si quantum devices, based on ^{31}P qubits are a rapidly maturing quantum technology with great promise, and should enable the extremely long lifetimes of electron and nuclear spins be harnessed in a scalable fashion. The two established fabrication techniques, i.e. ion-implantation and STM-assisted lithography, have so far enabled impressive demonstrations of rapid one-, two-, and three-qubit logic gates with high fidelity. Yet, scaling to several qubits and eventually a large-scale quantum processor remains a challenge due to the requirements of precisely controlled, efficient, and reproducible dopant placement in a low-noise qubit environment. Long-range qubit coupling schemes such as the electric dipole–dipole interaction may alleviate the requirement for precise donor placement but are yet to be experimentally realized. If proven viable, further coupling of Si:P qubits to microwave resonators may however present a path toward the exciting prospect of hybrid quantum technologies.

Acknowledgements

This research was supported under the Singapore National Research Foundation (NRF) Fellowship (NRF-NRFF2017-11) ‘Atomic Manipulations in 2D Materials for Quantum Computing Applications’ with further support from the Singapore Ministry of Education (MOE) Academic Research Fund Tier 3 Grant (MOE2018-T3-1-002). BW and KEJG are supported by the National Singapore Research Foundation (NRF) Competitive Research Programme NRF-CRP21-2018-0001. KEJG acknowledges funding support from Agency for Science, Technology and Research (#21709). KEJG, BW and TSK acknowledge support from a Singapore National Research Foundation Grant (NRF2021-QEP2-02-P07).

6. Color centers in wide-bandgap materials for quantum applications

Zhao Mu¹, Christoph Becher^{2,3} and Weibo Gao^{1,3,4}

¹ Division of Physics and Applied Physics, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371, Singapore

² Fachrichtung Physik, Universität des Saarlandes, Campus E2.6, 66123 Saarbrücken, Germany

³ The Photonics Institute and Centre for Disruptive Photonic Technologies, Nanyang Technological University, Singapore 637371, Singapore

⁴ Centre for Quantum Technologies, National University of Singapore, Singapore

Status

Color centers in semiconductor materials have been intensively studied since the first report of single negatively charged nitrogen vacancy (NV^-) color centers in diamond [68]. During the past two decades, their family members have grown from diamond NV^- centers to group IV split vacancy centers in diamond, Si vacancy (V_{Si^-}) and divacancy (VV^0) centers in silicon carbide (SiC), G-centers in Si, and the emerging V_B^- centers in hexagonal boron nitride (hBN) [69]. Most of these defects have well-known molecule configurations and clear energy structures thanks to various experimental characterization methods and density functional theory (DFT) calculations. These defects have distinct spin, optical, and charge properties, and additional functionality provided by their host materials along with nanofabrication. Exploiting these properties has enabled quantum sensing, quantum computation, and quantum communication.

Among the color center pool, the diamond NV^- center is the most studied system for quantum sensing and quantum network applications [68]. The diamond NV^- spin has a long coherence time and is a good sensor for the magnetic field, electrical field, temperature, and pressure. In particular, this center is most suitable for magnetic field sensing. The nanoscale NV^- quantum sensor leads to many new applications in biology and condensed matter physics [70]. The DC and AC magnetic field sensitivity has been pushed to the pT/\sqrt{Hz} range [71] and submillihertz spectral resolution has been achieved [72]. With the hyperpolarization between NV^- electron spins and target sample nuclear spins, high-resolution nuclear magnetic resonance (NRM) detection of a single cell has been reported [73]. The long spin coherence and the rich nuclear resources surrounding NV^- centers have enabled a quantum processor of ten qubits with coherence time up to minutes [74]. Quantum error correction and fault-tolerant operation have also been realized, promising future quantum computation. A quantum network composed of three nodes [75] was demonstrated after the realization of the deterministic entanglement of two nodes.

Two major issues for diamond NV^- centers are their low Debye–Waller factor [76] and their visible emission wavelength, making it challenging to realize long-distance key distribution. The group IV split vacancy centers with inversion symmetry, reducing their susceptibility to external fields, and a high Debye–Waller factor could be a potential candidate to realize quantum networks [77]. Recently, a two-qubit register (a negatively charged silicon vacancy (SiV^-) electron spin and a nearby ^{13}C nuclear spin) was constructed, coupled efficiently to a diamond nanocavity. This efficient nanophotonic interface was further utilized for memory-enhanced quantum communications, a cornerstone of long-distance key distribution [78]. Beyond this, heterogeneous on-chip integration of a diamond array containing group IV vacancies into an aluminum nitride photonic integrated circuit has been realized which may promise large-scale quantum information processing [79].

Apart from the diamond emitters, emitters in SiC benefit from the host material which could be grown at a large scale, easily doped, and nanofabricated into high quality factor nanostructures [80]. Taking these advantages, the emission wavelength of VV^0 centers can be tuned over terahertz ranges and the emission linewidth can reach almost the lifetime limit when using charge depletion. The VV^0 defects also exhibit excellent spin properties of 5 s coherence time and can realize single-shot readout [81]. In addition to this progress, the two-photon interference experiment and electrical readout of V_{Si^-} spin states have been realized. Moreover, SiC is a good host of various transition metal ions, among which the single vanadium (V) defect with emissions in the telecom range has been isolated. This progress demonstrates SiC defects as a promising platform for realizing quantum networks or quantum computation.

Besides these defects in bulk hosts, many kinds of optically active spin defects in hBN have been reported. Among them, the V_B^- center has been intensively investigated, enabling the identification of its molecular structure, integration into nanostructure, and application in sensing in various fields [82–84].

Current and Future Challenges

A common problem for both diamond emitters and SiC emitters for practical applications is the low photon extraction efficiency due to total internal reflection. This deficiency limits both the entanglement rates and the entanglement distribution distance, as well as the sensitivity of quantum sensors. The current solution to

Table 2. Optical and spin properties of color centers in diamond, SiC, and hBN.

	Single emitter generation	ZPL (nm)	LT DWF (%)	Lifetime limited linewidth	Two photon interferences	Spin	RT ODMR contrast (%)	T_2^b	References
Diamond	NV ⁻	637	3	Y	Y	1	30	>1 ms (300 K)	[85, 86]
	SiV ⁻	738	>70	Y	Y	1/2		0.3 ms (0.1 K)	[87, 88]
	GeV ⁻	602	61	Y	Y	1/2			[89, 90]
	SnV ⁻	620	57	Y		1/2		28.3 μs (1.7 K)	[91–93]
	PbV ⁻	550–554				1/2			[94]
3C-SiC	VV ⁰	1106	7			1	7.5 ^a	0.9 ms (20 K)	[95]
	N _C V _{Si} ⁻	1289	6			1			[96]
4H-SiC	V _{Si} ⁻	862 (V1)	40	Y (V1)	Y (V1)	3/2		83 μs (4 K)	[5, 97]
		858 (V1')							
		917 (V2)	8–9	Y (V2)			1 (V2)	0.85 ms (10 K)	
6H-SiC	VV ⁰	1038–1132	5			1	5–22	1.2 ms (kk) (20 K)	[95, 98]
	N _C V _{Si} ⁻	1176–1240				1	0.1	17 μs (hh) (300 K)	[99]
	V ⁴⁺	1278(h), 1335(k)	25 50			1/2			[100]
	V _{Si} ⁻	865 (V1)				3/2			[97]
h-BN		887 (V2)					0.03 (V3)		
		907 (V3)							
	VV ⁰	1093–1139				1	2	5.6 μs (QL2) (300 K)	[101]
	N _C V _{Si} ⁻	1155–1242				1			[102]
	V ³⁺	1309 (h)	45			1/2			[100]
	1352 (kl)	50							
	1388 (K2)	40							
		773				1	40		[103, 104]

LT: low temperature; RT: room temperature.

^a The contrast is obtained under the pulsed ODMR at 20 K.

^b The coherence time is estimated with the Hahn echo.

relax this limitation is to integrate color centers into nanostructured hosts [105], e.g. nanobeam cavities. However, these nanostructures often adversely impact the optical properties of color centers, i.e. their emission linewidths might be broadened by several to tens of times due to spectral diffusion created by surface defects introduced during nanofabrication. Moreover, different emitters mostly exhibit different emission wavelengths due to different *in-situ* environments. This broadened emission and environment dependent emission wavelength creates a barrier that must be overcome to achieve fully chip-integrated components for the quantum internet.

For building up a fiber-linked quantum network, both diamond NV^- and group IV split vacancy centers have their challenges besides their visible emission wavelength. The NV^- centers have a small Debye–Waller factor (3%) and an environment-sensitive emission, suggesting a limited photon emission rate and imposing a limited qubit operation speed. The SiV^- in diamond has a Debye–Waller factor of 70% which is beneficial for coherent photon generation (table 2). Nevertheless, the ground state spin coherence time is limited by the phonon bath which couples the ground state orbital doublet leading to the requirement of operating temperatures below 100 mK [87]. Despite these shortcomings, in the field of quantum networks the state of the art is set by NV^- centers [75].

As a key candidate in quantum sensing, the main challenge for diamond NV^- centers is the shortened coherence time and unstable charge state due to fluctuating charges when the NV^- centers are placed near the surface [106, 107]. This makes it challenging to sense biological samples which require nanodiamond hosts or for weak signal detection which needs a small, typically nanometer-scale, sensor–target distance. The sensitivity of NV^- center-based quantum sensors strongly depends on the density of the centers and on the spin coherence time. These two properties commonly trade off, i.e. too high densities lead to reduced spin coherence due to spin–spin interactions. It is thus important to answer the questions including how to get high-density diamond NV^- samples and how to get rid of the interaction limitation. On the other hand, SiC emitters seem to be potential sensor candidates given that VV^0 centers exhibit long coherence time and high optically-detected magnetic resonance (ODMR) contrast, and the SiC host material is also biocompatible. However, the generation of these defects in SiC nanoparticles has rarely been studied. The V_B^- centers in hBN are good sensors in terms of their high ODMR contrast and their 2D nature, but their short coherence time would limit their applications in AC field sensing. Moreover, the V_B^- centers exhibit low quantum yield, making it difficult to isolate a single V_B^- center to achieve single spin sensing. Unlike the V_B^- center, the structural composition of other defects is unclear, making it difficult or even impossible for a targeted or deterministic creation.

Advances in Science and Technology to Meet Challenges

Optical transitions of V_{Si}^- emitters in SiC and SiV^- emitters in diamond are predicted to be insensitive to the electrical field to the first order. The broadened emission of diamond and SiC emitters embedded into nanostructures are not intrinsic but due to the imperfect engineering process. A lifetime-limited emission linewidth is then expected by optimizing the fabrication recipe or exploiting the charge depletion method. Recently, negatively charged tin vacancy SnV^- centers in diamond nanostructures (nanopillars and waveguides) [91] and V_{Si}^- centers in reactive ion etching developed SiC waveguides were shown to exhibit lifetime limited emission [5]. Exploiting the p-i-n SiC junction, the charges surrounding the VV^0 centers are depleted, resulting in a 50 times narrower emission linewidth [108]. Though these techniques or special treatments may alleviate or resolve the spectral diffusion problem, the definitive solution still relies on the understanding of the underlying mechanism.

Once the spectral diffusion problem is under control, the color center emission into the zero phonon line (ZPL) and the overall emission rates can be enhanced by overlapping the cavity resonance wavelength and the color center emission wavelength, as demonstrated for all the above-mentioned emitters in diamond and SiC. Regarding a single quantum node, the spin–photon entanglement rates can be increased by several orders of magnitude thanks to the Purcell enhancement of spontaneous emission and the potentially higher repetition rates. With regard to different nodes, the spin–spin entanglement requires indistinguishable photons emitted from separated nodes. Besides improvement in defect center generation technologies, in particular high-temperature annealing to reduce inhomogeneous broadenings [90], this requires an additional degree of freedom for tuning the emission wavelength which is provided by strain engineering for group IV split vacancy centers, and stark shift for color centers without inversion symmetry. By overlapping the cavity resonance frequency and color center frequency, both on-chip quantum processors of several electron qubits and entanglement among multi-nodes can be conceived.

To achieve millisecond coherence time in cryogenic temperature for group IV split vacancy centers, the key is to decouple the ground orbital states from the phonons by augmenting the energy splitting between the ground doublet states. The strain engineering allows the ground state splitting to increase by ten times for SiV^- centers and prolonged the spin coherence time by a factor of six [109]. However, this benefit comes

with the cost of breaking down the inversion symmetry. Alternatively, the SnV^- and negatively charged lead vacancy PbV^- in diamond with greater splitting are potential choices. The SnV^- exhibits a spin coherence time of 0.3 ms at 1.7 K [92], about five times longer than the SiV^- coherence time at 100 mK (table 2).

To realize high-sensitivity (spatial or spectral) detection of targets, the following factors are of great importance: the coherence time, the number of the spins within a given volume and the sensor–target distance. Looking back through the history of diamond NV^- centers, several approaches have been used to prolong the coherence like surface engineering, dynamical decoupling, clock transition, and coherent driving of surface electrons. However, these techniques still could not push the coherence to the T_1 limited level. A comprehensive study of the decoherence sources with a complete toolbox to take care of them is still needed. Recently, diamond samples have been produced with high density of NV^- centers for which the spin–spin interactions begin to play limiting roles. Utilizing Hamiltonian engineering to overcome the undesired effects including interaction between NV^- centers, local disorders, and control errors, the authors demonstrated quantum sensing surpassing the interaction limit with this diamond NV^- sample [110–114]. This technique would benefit the development of magnetometers with higher sensitivity. hBN has been widely used to encapsulate 2D materials. The target–sensor distance thus could be brought within the subnanometer range. Naturally, the 2D sensor could play a complementary role for specific applications like sensing the magnetic field generated by 2D magnets.

The rich experience gained from studying the SiC and diamond color centers, along with the ongoing data mining assisted defect prediction, would benefit the discovery of more defects that may outperform the current defect platforms or would be more suitable for certain applications [115]. Meanwhile, these valuable resources would accelerate the study of emerging defects like hBN color centers and benefit the optimization of the optical and spin properties of the defects.

Concluding Remarks

The current color center platforms are not yet perfect. It is still hard to predict whether one platform will deliver a solution for all applications in the end or whether different defects will turn out to be suited best for specific tasks. The class of color centers host is expanding from 3D materials to 2D materials, and from wide-bandgap hosts to small-bandgap hosts. New color center discovery is also expected to be accelerated with the help of data-mining-based prediction. With increasing experience in the study of diamond and SiC color centers, both the optical and spin properties of the existing platforms improve with the combined efforts of materials engineering, enriched quantum control toolbox, and matured nanofabrication techniques. This deep understanding of the pros and cons of the current platforms, in turn, benefits the goal of designing quantum defects for specific applications. Moreover, it is worth noting the complementary roles played by hybrid systems including nanomechanical and superconducting systems. The mechanical driving of NV^- centers and SiV^- centers in diamond, and V_{Si}^- and VV^0 in SiC, have been demonstrated. Nanomechanical resonators including diamond/3C–SiC cantilevers and optomechanical microresonators have also been realized [110–113]. The following efforts should be devoted to the realization of single phonon–single spin interaction by utilizing defects featuring strong strain orbital coupling and engineering high-quality nanomechanical resonators. Beyond this, one can conceive the phononic quantum networks with color centers.

To realize a quantum network, integrating multiple color centers into nanostructures is the key ingredient but has not been fully demonstrated. Past progress was mainly realized with SiV^- in diamond, though diamond nanofabrication is less matured compared to its SiC and Si counterparts. The current progress in VV^0 centers in SiC is attractive. The recent demonstration of the prolonged spin coherence via clock transition, enhanced emission into the ZPL, tunable emission wavelength, and single-shot readout of spin states could pave the way to spin–photon entanglement and even on-chip quantum processors benefiting from the matured SiC nanofabrication techniques.

The diamond NV^- spin sensor has achieved many successes in quantum sensing and quantum communication, mainly based on its long spin coherence. Investigation into single NV^- and ensemble NV^- sensor-based quantum sensing, including materials engineering, and Hamiltonian engineering, toward higher sensitivity and better spectral resolution, would benefit their application in fundamental research and single cell or molecular detection. While it is still the leading platform for sensing applications, other systems with particular properties may play complementary roles. VV^0 centers with clock transition may be favorable for Ramsey-type measurements while hBN hosted defects may be preferred for quantum sensing of 2D materials.

Acknowledgements

We acknowledge the Singapore National Research Foundation through QEP Grant (NRF2021-QEP2-01-P02, NRF2021-QEP2-03-P01, 2019–0643 (QEP-P2) and 2019–1321 (QEP-P3)) and Singapore Ministry of Education (MOE2016-T3-1-006 (S)).

7. 2D materials for quantum technologies

Jeng-Yuan Tsai and Qimin Yan

Department of Physics, Northeastern University, Boston, MA 02115, United States of America

Status

Being atomically thin, amendable by defects and interfaces, and sensitive to external stimuli, 2D materials offer a broad range of material properties to serve as functional device components for a set of emerging quantum technologies, including quantum computing, quantum communication, and quantum sensing (figure 8). Two-dimensional materials have demonstrated a variety of novel functionalities and been applied in integrated quantum devices (such as atomically thin quantum light-emitting diodes (LEDs), quantum-confined Stark effect modulators, and on-chip quantum photonic devices).

The development of solid-state qubit systems for quantum computation is crucial to the anticipated quantum revolution. Two-dimensional materials, including graphene, h-BN, and transition metal dichalcogenides (TMDs), have been explored as host materials in four qubit design strategies including QDs, defect spins, superconducting junction, and topological qubits [7].

As discussed in sections 5 and 6, point defects in their hosts (such as Si and wide-band-gap materials) may create coherent quantum states for quantum computing and communication. Compared to conventional bulk material systems, the planar structures of 2D materials present a superior platform for realizing controlled creation and manipulation of defect qubits for scalability. Several quantum defects in h-BN with triplet spin states, including the C–vacancy complex and negatively charged B vacancy [116], have been proposed as spin qubits. Antisite defects in TMDs have been identified as viable spin qubits (figure 9(a)) [117].

Using the spin states of trapped charges and the Coulomb blockade phenomenon, QD qubits can be created [118]. Coulomb-blockade behavior has been observed in graphene QD qubits and coupled multi-QD qubits have been proposed based on graphene nanoribbons. Graphene QDs have demonstrated great potential as photodetectors as well as nanoprobes for biosensing and bioimaging. Owing to spin–orbit interaction and unique valley physics, qubits based on TMD QDs, including spin qubits, valley qubits and hybrid spin–valley qubits, have been theoretically explored [119]. Coulomb blockade in encapsulated MoS₂ QDs [7] and WSe₂ QDs [120] has been observed. Solid-state qubits can also be realized using superconducting circuits based on Josephson junctions (JJs). The diverse properties of 2D materials provide the opportunity to fabricate all-2D JJ, eliminating device complexities.

Optical transitions created by deep defect states or defect-bound excitons in SPEs can be utilized for quantum communication techniques. In 2015, the first SPEs in 2D materials were demonstrated in monolayer WSe₂, a semiconductor compound in the TMD family [121, 122]. Later on, h-BN emerged as a promising material platform of polarized high-brightness SPEs that operate at room temperature [123]. The emission source in h-BN has been identified to be C-related. Going beyond individual 2D materials, quantum emitters have been observed in twisted 2D heterojunctions from moiré-trapped excitons [124].

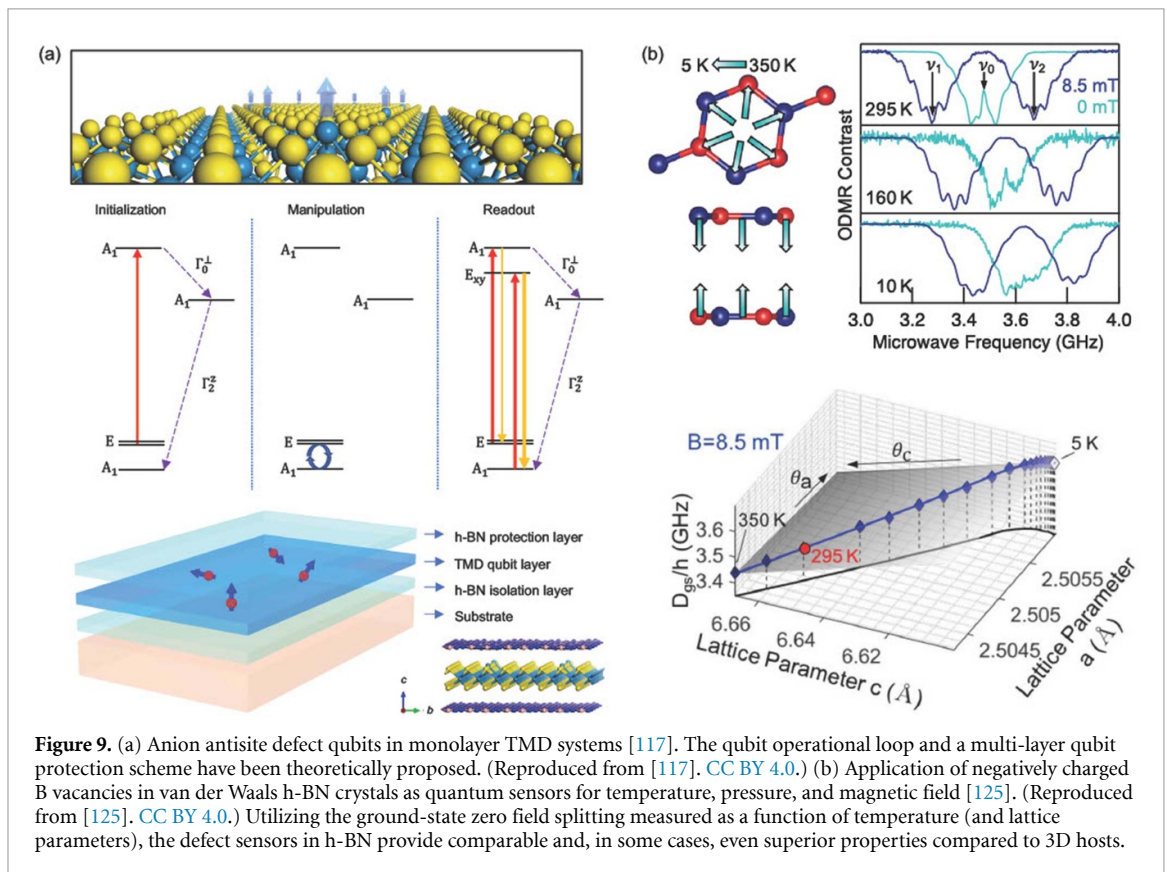
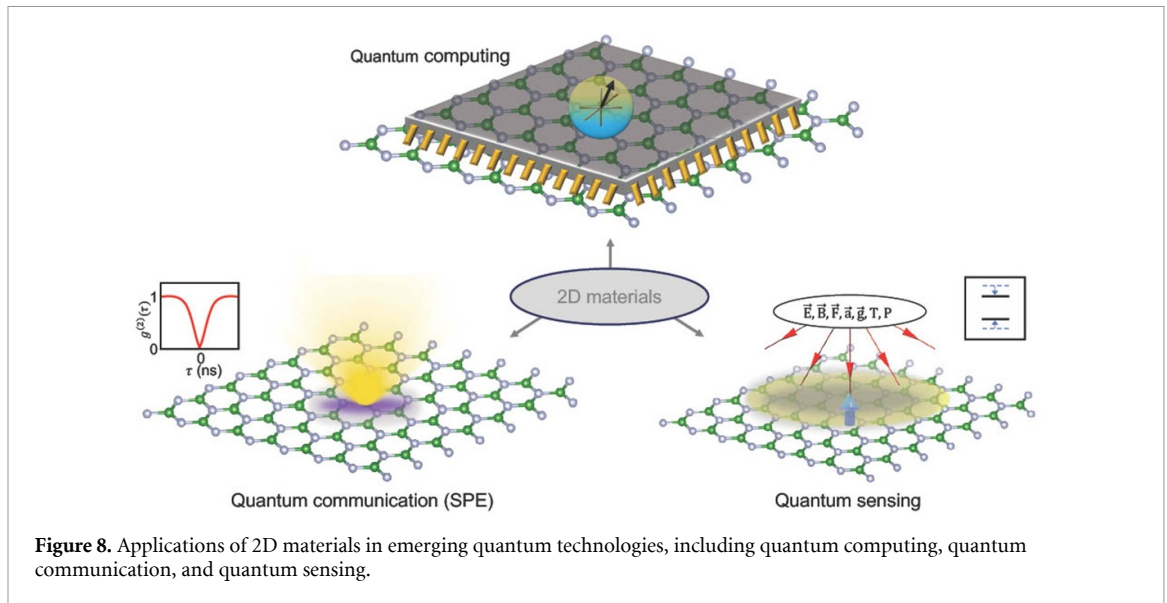
As the key component of quantum sensing technology, solid-state quantum sensors use atom-like systems, including defects in solids, with discrete and tunable energy levels to measure perturbations in the environment. While providing natural protection, the use of bulk materials limits the sensitivity and impedes on-chip integration. Two-dimensional materials with defects can greatly improve the versatility and sensitivity of quantum-sensing technologies. Recently, negatively charged B vacancies in h-BN have been identified as promising quantum sensors for temperature, pressure, and magnetic field (figure 9(b)) [125].

Current and Future Challenges

Despite its vast potential, the field is still in its early stage and full exploitation of 2D materials for quantum technologies is facing challenges that call for advances in science and technologies.

In the field of qubits in 2D materials, great challenges remain for qubit fabrication, characterization, and device integration. It is still a great challenge to create stable defect qubits in h-BN or TMDs in a controllable manner, which is important for deterministic qubit fabrication and scalability in the long run. Another challenge is to provide protection for defect qubits against environmental perturbation. For QD qubits, the imperfect edges of graphene create disorders that compromises quality and reproducibility even with h-BN encapsulation. The proposed spin and valley qubits in semiconducting TMD QDs are still waiting for experimental realization. Going forward, major long-term challenges are scaling up qubit circuits and realizing quantum computation based on coordinated quantum networks.

For SPEs in 2D materials, there are challenges to improve the performance of these emitters in terms of indistinguishability, room-temperature accessibility, electrical and photonic addressability, and entanglement. For instance, entangled light sources have not been demonstrated in any 2D materials yet. It is



challenging to unravel the origins of the defects that are responsible for the observed quantum emission in h-BN. It is also necessary to identify SPEs in other 2D materials that cover a broader spectrum (from near infrared to visible) for usage in quantum technologies such as optical-fiber-based quantum communication.

In the field of quantum sensing, the demonstration of B vacancy quantum sensors in multi-layer h-BN [125] is an encouraging starting point, but there is still a long way to go before it reaches the ultra-thin monolayer limit. The 2D nature of monolayer van der Waals materials is expected to be a double-edged sword for quantum sensing. With substantial charge or spin noise from the environment, there is still an unanswered question as to whether quantum defects in a specific charge state can exist in monolayers and what the sensing performance would be in that case. A long-term challenge is to identify quantum defects in other 2D materials in the single defect limit for nanoscale sensing.

In general, full exploitation of 2D materials for quantum technologies faces challenges in fabrication methods and large high-quality materials are pivotal to practical deployment of the 2D quantum platform. Another general challenge in the field is to go beyond the existing 2D materials.

Advances in Science and Technology to Meet Challenges

Moving forward, unprecedented advances in 2D material growth and characterization have been achieved, which will meet the major challenges in deploying 2D materials for quantum technologies.

Synthetic strategies and assembly schemes with wafer-scale homogeneity have been developed for 2D materials such as h-BN and TMDs. With the advances in material fabrication techniques, 2D materials can be interfaced with other materials to form multifunctional heterostructures, opening new possibilities for on-chip quantum communication and quantum sensing.

Defect fabrication in 2D materials can be improved by using low-energy ion implantation and bottom-up synthesis methods based on the achievements made in diamond. The developments in scanning tunneling and transmission electron microscopy provide the capability to image individual defects in 2D solids with atomic resolution. Rapid progress is being made in physical characterization of SPEs, such as super-resolution imaging and *ab initio* modeling of defect-based emission sources. The development of techniques to integrate SPEs in 2D quantum hosts with atomically thin lenses and metasurfaces for light manipulation will lead to integrated quantum nanophotonic networks and other quantum devices.

Ab initio computational methods associated with defect theory are being actively developed toward fundamental understanding of various aspects of quantum defect states [116, 126], such as excitations and relaxations, spin coherence, correlation effects, and intersystem crossing phenomena. Meanwhile, the continued development of various QD systems based on TMDs, superconducting 2D materials, and their heterojunctions will create multiple alternative and promising platforms for realizing diverse types of qubits for quantum technologies.

Another major advance associated with 2D materials is the fabrication of 2D heterostructures with functionalities unavailable in bulk materials. The continued development of encapsulation/insulation techniques by fabricating 2D heterojunctions will provide qubits and SPEs with protection against environmental effects. The understanding in strongly correlated physics and topology in quantum materials has enabled 2D heterojunction systems as a robust quantum simulation platform [127]. With the recent developments in synthetic 2D materials, the number of building blocks for heterojunction-based quantum systems will continue to grow.

Going beyond h-BN and TMDs, the development of data-driven material discovery approaches such as the construction of 2D material databases with defect information [128] will accelerate the search for other 2D material systems that enable integration in solid-state, on-chip quantum devices. With the aid of data-driven approaches [117, 128], the discovery of stable and coherent qubits and SPEs in novel 2D materials systems can bring a breakthrough to emerging quantum technologies, even at room temperature.

Concluding Remarks

Two-dimensional materials host inherent advantages toward the development of emerging quantum technologies, including quantum computing, quantum communication, and quantum sensing. The emergence of a diverse set of quantum phenomena in 2D materials offers great potential for developing integrated quantum technologies. In addition, heterostructures of 2D materials offer a new quantum design platform with functionalities unavailable in individual materials. Despite significant advances in 2D material synthesis and characterization, 2D quantum technologies are far from mature and major challenges remain. With the rapid progress in scientific and technological developments in the field, the future of 2D materials for quantum technologies is bright.

Acknowledgements

This work is supported by the National Science Foundation under Grant No. 2144936.

8. III–V optical active semiconductor QDs for quantum technologies

Tobias Huber-Loyola and Sven Höfling

Technische Physik, Universität Würzburg, Am Hubland, 97074 Würzburg, Germany

Status

Semiconductor QDs are sources of single [129] and entangled photons [130]. They combine the advantages of semiconductor fabrication technologies with the advantages of single atoms or ions. They possess a large dipole moment [131] for efficient light matter coupling [132] and individual electrons or holes can be used as local spin qubits [133]. In particular, their high rates of single photons, when embedded into micro- or nanoresonators [134], has made them one of the best choices for optical experiments with discrete variables.

Semiconductor QDs form as zero-dimensional objects in epitaxial growth, which either form through built-in strain or through hole etching and infilling. Their small scale, together with the dimensionality, leads to discrete energy levels which can be occupied by single electrons, holes, or excitons, which are a quasi-particle consisting of a bound electron–hole pair. The transition energy of a charge complex is given by the confinement potential, i.e. the size of the QD and its material composition. By varying the used group III or group V material, the barrier as well as the QD potential can be varied, leading to flexibility in the emission wavelength over a wide range of the spectrum.

Comparing them with individual atoms, they have the advantage of being in a solid-state semiconductor environment. This makes them easy to use since they do not need to be trapped with optical tweezers or electromagnetic fields. However, they still need to be cooled to cryogenic temperatures to minimize phonon coupling effects. Furthermore, their environment can be engineered using doping and by changing semiconductor compounds. Nevertheless, being in a semiconductor environment also brings challenges through coupling to the environment, which is intrinsic and thus can be only minimized to some extent but will never disappear completely.

An approach to using photons from QDs in quantum computing is to generate cluster-states of photons through interaction with local spins. There are proposals on how to extend this to higher-dimensional cluster states using singlet–triplet states in QD molecules [135], where first experimental results on controlling the system have been obtained [136].

Recent results used QDs as a source of indistinguishable photons to run a boson sampling experiment, which is a special purpose quantum computer. The experiments were able to enter a regime where the result cannot be computed easily on classical computers any longer [137].

Another recently published work shows that with proper material engineering the spin coherence times of QDs can be extended into the millisecond regime, which will allow exciting experiments in the future [138].

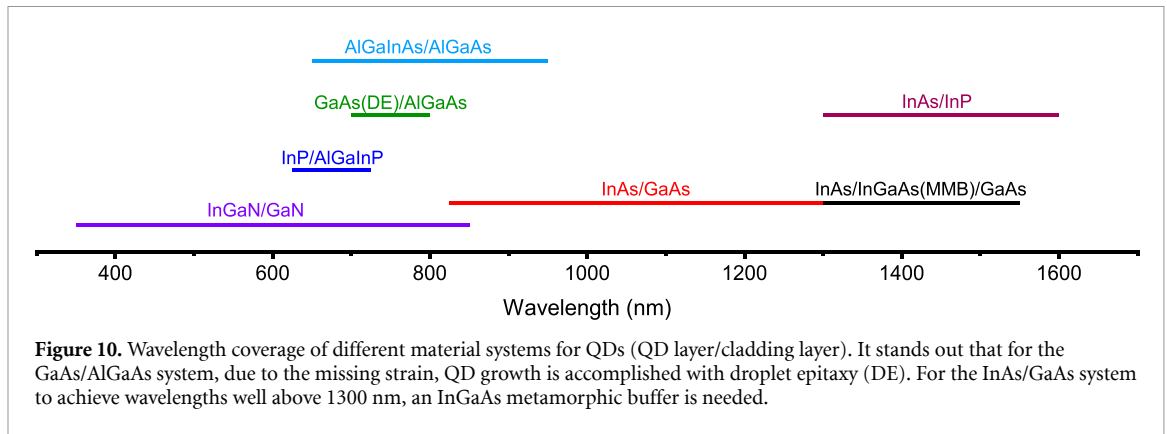
Current and Future Challenges

One ever-present challenge in QDs is their random growth. They grow randomly in size and spatial distribution, which requires post-growth selection of well-performing QDs. Attempts to grow site controlled QDs [139] have not yet been able to reproduce the excellent optical performance of self-assembled QDs. To gain higher device yields, extra tuning knobs, like electric fields [140], magnetic fields [141], or mechanical strain [142] can be integrated into the devices, which makes their fabrication more challenging.

Furthermore, the high refractive index of III/V semiconductors forces one to use nanophotonic waveguides [143] or resonators [144] so as not to suffer from bad extraction efficiencies. The resonator design is often in contradiction with the additional wanted tuning knobs, making photonic device design challenging.

Another challenge is inhomogeneous line broadening [145] which makes successively emitted photons partly distinguishable [146]. This broadening comes in general from interaction with a fluctuating environment, which is mostly given by charge noise. For instance, when etching cavities, one often introduces surface damage close to the QDs that increases their inhomogeneous linewidth due to fluctuating surface charge states. Therefore, increasing the distance to etched surfaces as well as the application of passivation layers will be important optimization routes for further improved figures of merit. In quantum technological relevant experiments with photons, the indistinguishability of photons is a limiting factor in two-qubit gate fidelity. This is currently also limiting the performance of the above mentioned boson sampler.

Recent works have shown that QDs can be grown such that they emit directly in the telecom C- (1530–1565 nm) or O-band (1260–1360 nm), which makes them excellent candidates for quantum communication since their photons will not need to go through an additional stage of frequency conversion (see also figure 10). However, these new materials bring their own challenges in growth and all their properties, including the photons as well as the spin properties, have to be investigated and understood.



Advances in Science and Technology to Meet Challenges

Recently, many groups have developed *in-situ* or two-step fabrication techniques to overcome the challenge of random growth by deterministic device fabrication. In these techniques, QDs are semi-automatically pre-selected, and nanostructures are fabricated around them. Device designs including diodes that are compatible with resonators and devices with strain-tuning nanophotonic devices to achieve tunability have been demonstrated. However, missing is the combination of the two, but this seems to be a technological effort that will be solved soon.

To tackle inhomogeneous broadening, several options are possible and maybe a combination of them will be required in the end. Optimizing growth techniques and material quality helps to minimize broadening from parasitic background doping and fluctuating charges in the vicinity of the QDs. Introducing diode structures can minimize electronic noise through Fermi pinning of the previously floating Fermi levels. Surfaces close to QDs can be treated with surface passivation such as S or O compounds to minimize the fluctuation of surface charges.

Concluding Remarks

QDs are promising candidates for the generation of complex highly entangled states due to their high photon generation rates and ease of integration into nanophotonic resonators. Furthermore, with future development on spin coherence times they offer an alternative for local memory, where source and memory can be in the same device.

Acknowledgements

Tobias Huber-Loyola acknowledges funding from the Federal Ministry of Education and Research (BMBF) through the Quantum Futur (FKZ: 13N16272) and funding from the German Research Foundation (DFG) in the project HU2985/1-1 and the German Israeli Project Cooperation (DIP) project FI 947/6-1. Sven Höfling acknowledges funding from BMBF through QR.X (FKZ: 16KISQ010), PhotonQ (FKZ: 13N15759) and MHLASQU (FKZ: 13N16027), and the DFG within the project HO5194/8-1 and the German Israeli Project Cooperation (DIP) project FI 947/6-1.

9. Superconducting materials for single photon detectors

Samuel Gyger, Stephan Steinhauer and Val Zwiller

KTH Royal Institute of Technology, Stockholm 10691, Sweden

Status

Quantum information processing has grown to extraordinary prominence within today's research and development environments, with superconducting materials playing a decisive role in the related device technologies. Immense progress in different fields has relied on continuous advances in our knowledge of superconducting structures at mesoscopic and nanoscopic scales, including their interactions with electromagnetic radiation from microwave to optical frequencies. To date, superconducting materials are key enablers for a variety of emerging quantum technologies with the potential for significant societal and commercial impact, ranging from superconducting quantum computing to single photon detectors.

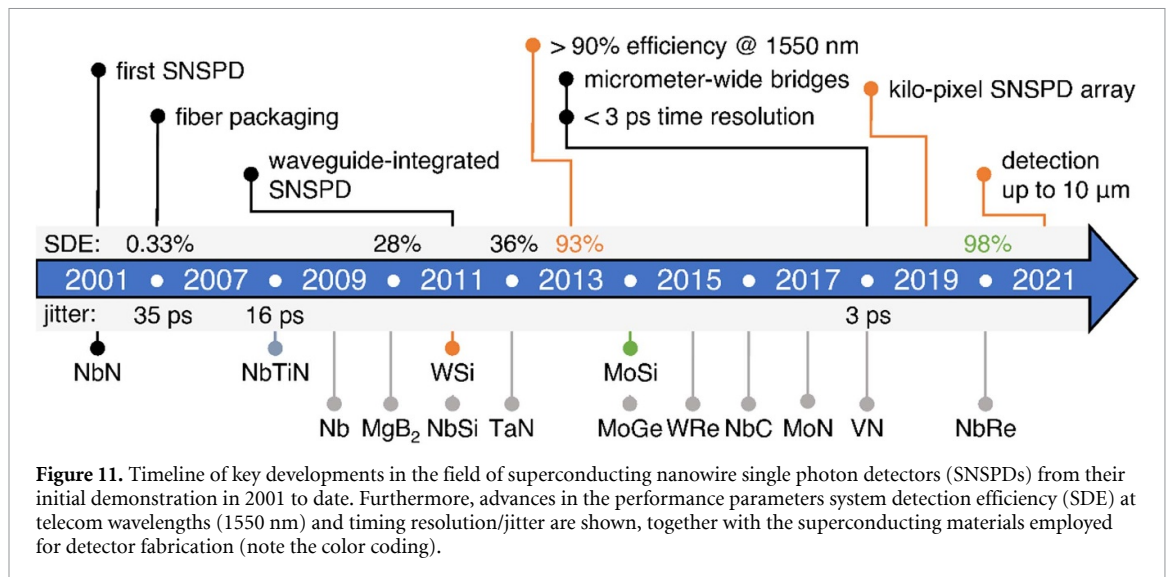
Research efforts over the last several decades have established that superconducting devices can offer unrivalled capabilities for the detection of single photons in the visible and near-infrared wavelength range. Based on the photon absorption-induced effects in superconducting materials, different operation principles have been elaborated, most notably kinetic inductance detectors, transition edge sensors and superconducting nanowire single photon detectors (SNSPDs). The latter technology, initially demonstrated in 2001 [147], provides exciting prospects for a wide range of applications in modern photonic quantum technologies due to the unique combination of close-to-unity detection efficiencies exceeding 98% [148, 149], high time resolution below 3 ps [150], extremely low noise/dark counts below the millihertz range [151], photon number resolution capabilities [152, 153], possibility for integration with photonic circuits [154], and suitability for operation in compact cryocoolers and comparatively simple readout schemes [8, 155]. In this roadmap perspective, we hence focus on SNSPD technology and related materials science aspects as well as challenges, providing an outlook on future directions. Note that, recently, microwire-based superconducting devices exhibiting single photon detection capabilities have been obtained, relying on comparable operation principles.

Previous advances in single photon detectors based on SNSPDs are summarized in figure 11, presenting a timeline of technological milestones. The relevant superconducting materials and their first uses in detector fabrication have been compiled, together with selected performance improvements in terms of system detection efficiency (SDE) at telecom wavelengths (1550 nm) and timing resolution/jitter. While the significant evolution in the field has also been based on progress in nanofabrication, photonic packaging, measurement technology and cryogenics, the growth of optimized superconducting materials with tailored properties has certainly played a vital role in these developments which cannot be underestimated.

Current and Future Challenges

The fabrication of SNSPD devices relies on the deposition of superconducting thin films with thicknesses around 10 nm and below. The growth process is linked with considerable requirements on homogeneity and process control, as the device properties and performance crucially rest on various, often inter-dependent, material parameters, such as the superconducting gap, scattering rates for electrons and phonons, crystalline structure, level of disorder/defects, thickness and morphology. During the last two decades, magnetron sputtering has served as workhorse for the fabrication of superconducting thin films employed for SNSPD devices; if not mentioned otherwise, the prior reports reviewed here have relied on this deposition technique. The superconducting materials can be broadly divided into two classes, i.e. polycrystalline and amorphous thin films. The former (typically nitrides) commonly exhibit superior time resolution and short dead times, whereas the latter (typically silicides) have been successfully employed in high-yield device fabrication.

The substantial evolution in SNSPD device performance and functionality has progressed along several fronts: the detector SDE has almost reached its fundamental limit of unity efficiency, while further optimization will most probably rely on improving optical coupling and light absorption in the detector structure, at least in the visible and near-infrared wavelength range. For the detection of long-wavelength photons in the infrared, superconductors with a small superconducting gap and/or reduced free carrier density are beneficial [156]—a challenge that can be addressed by the choice of material and stoichiometry optimization. On the other hand, the physical limitations on achieving the highest timing resolution/lowest jitter have remained elusive, with the current record value reported being below 3 ps. While multiple factors contribute to the overall detector jitter (e.g. electronic noise, signal propagation delay), it has been proposed that inhomogeneity of the superconducting material as well as its inelastic scattering time contribute to the intrinsic jitter [157]. In this regard, engineering superconducting device structures that minimize intrinsic jitter constitute a major challenge for realizing SNSPDs with optimized timing resolution. To further boost the functionality of SNSPD-based detector technology, it will be required to massively increase the active area



and the number of pixels, for instance for single-photon imaging and large-scale photonic integrated circuits. Recent progress [158] has resulted in arrays with more than 1000 pixels and active areas approaching 1 mm^2 , which still falls significantly short of other single-photon detector technologies. From a materials science point of view, the major challenge connected to SNSPD up-scaling is the limited homogeneity of the superconducting thin films employed for device fabrication. Moreover, to address challenges related to the cryogenic cooling system (cooling power budget at a given base temperature versus heat load per channel), it would be particularly valuable to fully fathom the optimum boundary conditions of available superconducting materials. In this regard, it is also a major challenge to develop new materials suitable for detector operation at higher temperatures (for current devices most commonly below 3 K).

Advances in Science and Technology to Meet Challenges

Future innovation will critically depend on the availability of specifically optimized superconducting materials, obtained either via refining widely used magnetron sputtering processes or via establishing alternative growth techniques. It will be of utmost importance to push the limits of currently prevalent polycrystalline and amorphous thin film materials further, as well as to gain more insight into the performance differences of these two material classes. Relying on an alternative growth technique, epitaxial NbN thin films (thickness 7 nm) on AlN-on-sapphire substrates have been achieved with molecular beam epitaxy (MBE), resulting in single crystalline layers with the NbN {111} axis aligned with the AlN *c*-axis [159]. Narrow nanowire linewidths down to 20 nm were required to approach saturated internal detection efficiency in the near-infrared. Nevertheless, this report confirmed the potential of MBE-grown films, as they showed high critical temperatures as well as lower sheet resistance and considerably higher critical current density compared to their polycrystalline counterparts. Further research will be needed to elaborate how single crystalline films with such a low degree of disorder can be optimally utilized for single photon detectors and if they can provide advantages in terms of homogeneity for high-yield fabrication. Another avenue toward the latter is the use of plasma-enhanced atomic layer deposition (PEALD) for superconducting material growth. In general, atomic layer deposition (ALD) is considered a deposition method well suited for achieving layers with excellent uniformity that are pinhole-free and conformal to the substrate, which is highly desirable from an SNSPD perspective. The viability of growing NbN thin films with PEALD has been shown, resulting in prospects for large-scale fabrication and for achieving detection capabilities further into the infrared [160].

In addition, future advances could be a result of innovation linked with the use of alternative superconducting material systems. A notable example in this regard is MgB₂, which exhibits high critical temperatures above 30 K and low kinetic inductance (30–60 times lower compared to NbN). High-quality MgB₂ thin films (thickness 5 nm) have been achieved with hybrid physical–chemical vapor deposition and SNSPDs with only 130 ps relaxation time have been obtained [161], making such devices ideally suited for operation at high photon count rates. Other research efforts have been directed toward developing SNSPD devices with cuprate high-temperature superconductors, which are particularly appealing due to the high critical temperatures above the liquid N boiling point. Major challenges still lie ahead, ranging from solving technological difficulties (e.g. material instability, patterning processes) to unveiling the physical mechanisms and operation conditions for single photon detection. However, recent results on YBa₂Cu₃O_{7-x} nanowires,

demonstrating hotspot formation and phase slip control induced by single photons, hold great promise for the successful realization of devices with superior timing resolution in the future [162].

Concluding Remarks

Advances in understanding and controlling superconducting material properties at the nanoscale have played a key role in achieving SNSPD devices with superior performance and their burgeoning commercialization. To guide future developments, it will be crucial to identify structure-property relationships of the relevant superconducting materials, correlating parameters such as (poly)crystalline/amorphous structure, chemical composition and defect density with transport characteristics and SNSPD performances. Studying the employed materials and devices from a materials science point of view will not only deepen our insights into the fundamental operation principles but also enable technological advances, including large detector arrays for biological and astronomical imaging as well as single photon spectroscopy providing a time resolution approaching the capabilities of streak cameras. This will give us the opportunity to reconcile successful detector concepts with promising material systems, specifically geared toward realizing SNSPD devices that simultaneously combine close-to-ideal performance metrics across all relevant figures of merit.












Acknowledgements

We acknowledge the support from the European Union's Horizon 2020 Research and Innovation Action under Grant Agreement Nos. 899824 (SURQUID) and 899580 (FastGhost).

Data availability statement

No new data were created or analyzed in this study.

ORCID iDs

Christoph Becher  <https://orcid.org/0000-0003-4645-6882>
Swastik Kar  <https://orcid.org/0000-0001-6478-7082>
Christian D Marciniak  <https://orcid.org/0000-0001-8401-3981>
Thomas Monz  <https://orcid.org/0000-0001-7410-4804>
John G Bartholomew  <https://orcid.org/0000-0003-0780-2471>
Huanqian Loh  <https://orcid.org/0000-0002-7325-0815>
Teck Seng Koh  <https://orcid.org/0000-0002-4030-8232>
Bent Weber  <https://orcid.org/0000-0001-8586-127X>
Samuel Gyger  <https://orcid.org/0000-0003-2080-9897>
Stephan Steinhauer  <https://orcid.org/0000-0001-6875-6849>
Val Zwiller  <https://orcid.org/0000-0002-5726-1063>

References

- [1] Pino J M *et al* 2021 Demonstration of the trapped-ion quantum CCD computer architecture *Nature* **592** 209–13
- [2] Kindem J M, Ruskuc A, Bartholomew J G, Rochman J, Huan Y Q and Faraon A 2020 Control and single-shot readout of an ion embedded in a nanophotonic cavity *Nature* **580** 201–4
- [3] Đorđević T, Samutpraphoot P, Ocola P L, Bernien H, Grinkemeyer B, Dimitrova I, Vuletić V and Lukin M D 2021 Entanglement transport and a nanophotonic interface for atoms in optical tweezers *Science* **373** 1511–4
- [4] Bussmann E, Butera R E, Owen J H G, Randall J N, Rinaldi S M, Baczewski A D and Misra S 2021 Atomic-precision advanced manufacturing for Si quantum computing *MRS Bull.* **46** 607–15
- [5] Babin C *et al* 2022 Fabrication and nanophotonic waveguide integration of silicon carbide colour centres with preserved spin-optical coherence *Nat. Mater.* **21** 67–73
- [6] Schimpf C, Reindl M, Basso Basset F, Jöns K D, Trotta R and Rastelli A 2021 Quantum dots as potential sources of strongly entangled photons: Perspectives and challenges for applications in quantum networks *Appl. Phys. Lett.* **118** 100502
- [7] Liu X and Hersam M C 2019 2D materials for quantum information science *Nat. Rev. Mater.* **4** 669–84
- [8] Esmaeil Zadeh I, Chang J, Los J W N, Gyger S, Elshaari A W, Steinhauer S, Dorenbos S N and Zwiller V 2021 Superconducting nanowire single-photon detectors: a perspective on evolution, state-of-the-art, future developments, and applications *Appl. Phys. Lett.* **118** 190502
- [9] Giustino F *et al* 2020 The 2021 quantum materials roadmap *J. Phys. Mater.* **3** 042006
- [10] Burkard G, Ladd T D, Nichol J M, Pan A and Petta J R 2021 Semiconductor spin qubits (arXiv:2112.08863)
- [11] Adcock J C, Bao J, Chi Y, Chen X, Bacco D, Gong Q, Oxenløwe L K, Wang J and Ding Y 2021 Advances in silicon quantum photonics *IEEE J. Sel. Top. Quantum Electron.* **27** 6700224
- [12] Chen S, Raha M, Phenicie C M, Ourari S and Thompson J D 2020 Parallel single-shot measurement and coherent control of solid-state spins below the diffraction limit *Science* **370** 592
- [13] Ahlefeldt R, Hush M and Sellars M 2016 Ultranarrow optical inhomogeneous linewidth in a stoichiometric rare-earth crystal *Phys. Rev. Lett.* **117** 250504

- [14] Brown K R, Chiaverini J, Sage J M and Häffner H 2021 Materials challenges for trapped-ion quantum computers *Nat. Rev. Mater.* **6** 1–14
- [15] Bruzewicz C D, Chiaverini J, McConnell R and Sage J M 2019 Trapped-ion quantum computing: progress and challenges *Appl. Phys. Rev.* **6** 021314
- [16] Blatt R and Wineland D 2008 Entangled states of trapped atomic ions *Nature* **453** 1008–15
- [17] Romaszko Z D, Hong S, Siegele M, Puddy R K, Lebrun-Gallagher F R, Weidt S and Hensinger W K 2020 Engineering of microfabricated ion traps and integration of advanced on-chip features *Nat. Rev. Phys.* **2** 285–99
- [18] Monroe C and Kim J 2013 Scaling the ion trap quantum processor *Science* **339** 1164–9
- [19] Allcock D T C, Campbell W C, Chiaverini J, Chuang I L, Hudson E R, Moore I D, Ransford A, Roman C, Sage J M and Wineland D J 2021 omg blueprint for trapped ion quantum computing with metastable states *Appl. Phys. Lett.* **119** 214002
- [20] Ahlefeldt R L, McAuslan D L, Longdell J J, Manson N B and Sellars M J 2013 Precision measurement of electronic ion-ion interactions between neighboring Eu $3+$ optical centers *Phys. Rev. Lett.* **111**
- [21] Thiel C W, Böttger T and Cone R L 2011 Rare-earth-doped materials for applications in quantum information storage and signal processing *J. Lumin.* **131** 353
- [22] Goldner P, Ferrier A and Guillot-Noël O 2015 Rare earth-doped crystals for quantum information processing *Handbook on the Physics and Chemistry of Rare Earths* vol 46, ed J-C G Bünzli and V K Pecharsky (Elsevier: Amsterdam) pp 1–78
- [23] Zhong M, Hedges M P, Ahlefeldt R L, Bartholomew J G, Beavan S E, Wittig S M, Longdell J J and Sellars M J 2015 Optically addressable nuclear spins in a solid with a six-hour coherence time *Nature* **517** 177
- [24] Afzelius M and de Riedmatten H 2015 Quantum light storage in solid state atomic ensembles *Engineering the Atom-Photon Interaction* ed A Predojević and M W Mitchell (Switzerland: Springer)
- [25] Chanelière T, Hétet G and Sangouard N 2018 Quantum optical memory protocols in atomic ensembles *Advances In Atomic, Molecular, and Optical Physics* vol 67 (Amsterdam: Elsevier) pp 77–150
- [26] Zhong T and Goldner P 2019 Emerging rare-earth doped material platforms for quantum nanophotonics *Nanophotonics* **8** 2003
- [27] Ferrenti A M, de Leon N P, Thompson J D and Cava R J 2020 Identifying candidate hosts for quantum defects via data mining *NPJ Comput. Mater.* **6** 126
- [28] Saglamyurek E, Sinclair N, Jin J, Slater J A, Oblak D, Bussières F, George M, Ricken R, Sohler W and Tittel W 2011 Broadband waveguide quantum memory for entangled photons *Nature* **469** 512–5
- [29] Xia K et al 2022 Tunable microcavities coupled to rare-earth quantum emitters *Optica* **9** 445–50
- [30] Yin C, Rancic M, de Boo G G, Stavrias N, McCallum J C, Sellars M J and Rogge S 2013 Optical addressing of an individual erbium ion in silicon *Nature* **497** 91–94
- [31] Weiss L, Gritsch A, Merkel B and Reiserer A 2021 Erbium dopants in nanophotonic silicon waveguides *Optica* **8** 40
- [32] Casabone B, Deshmukh C, Liu S, Serrano D, Ferrier A, Hümmer T, Goldner P, Hunger D and de Riedmatten H 2021 Dynamic control of Purcell enhanced emission of erbium ions in nanoparticles *Nat. Commun.* **12** 3570
- [33] Corrielli G, Seri A, Mazzera M, Osellame R and de Riedmatten H 2016 Integrated optical memory based on laser-written waveguides *Phys. Rev. Appl.* **5** 054013
- [34] Serrano D, Kuppusamy S Kumar, Heinrich B, Fuhr O, Hunger D, Ruben M and Goldner P 2022 Ultra-narrow optical linewidths in rare-earth molecular crystals *Nature* **603** 241–6
- [35] Ebadi S et al 2021 Quantum phases of matter on a 256atom programmable quantum simulator *Nature* **595** 227–32
- [36] Barredo D, Lienhard V, de Léséleuc S, Lahaye T and Browaeys A 2018 Synthetic three-dimensional atomic structures assembled atom by atom *Nature* **561** 79–82
- [37] Browaeys A and Lahaye T 2020 Many-body physics with individually controlled Rydberg atoms *Nat. Phys.* **16** 132–42
- [38] Bernien H et al 2017 Probing many-body dynamics on a 51atom quantum simulator *Nature* **551** 579–84
- [39] de Léséleuc S, Lienhard V, Scholl P, Barredo D, Weber S, Lang N, Büchler H P, Lahaye T and Browaeys A 2019 Observation of a symmetry-protected topological phase of interacting bosons with Rydberg atoms *Science* **365** 775–80
- [40] Brown M O, Thiele T, Kiehl C, Hsu T - W and Regal C A 2019 Gray-molasses optical tweezer loading: controlling collisions for scaling atom-array assembly *Phys. Rev. X* **9** 011057
- [41] Aliyu M M, Zhao L, Quek X Q, Yellapragada K C and Loh H 2021 D1 magic wavelength tweezers for scaling atom arrays *Phys. Rev. Res.* **3** 043059
- [42] Schymik K - N, Pancaldi S, Nogrette F, Barredo D, Paris J, Browaeys A and Lahaye T 2021 Single atoms with 6000-second trapping lifetimes in optical-tweezer arrays at cryogenic temperatures *Phys. Rev. Appl.* **16** 034013
- [43] Madjarov I S, Covey J P, Shaw A L, Choi J, Kale A, Cooper A, Pichler H, Schkolnik V, Williams J R and Endres M 2020 High-fidelity entanglement and detection of alkaline-earth Rydberg atoms *Nat. Phys.* **16** 857–61
- [44] Cairncross W B, Zhang J T, Picard L R B, Yu Y, Wang K and Ni K - K 2021 Assembly of a rovibrational ground state molecule in an optical tweezer *Phys. Rev. Lett.* **126** 123402
- [45] Kim M E, Chang T-H, Fields B M, Chen C-A and Hung C-L 2019 Trapping single atoms on a nanophotonic circuit with configurable tweezer lattices *Nat. Commun.* **10** 1647
- [46] Covey J P, Sipahigil A, Szoke S, Sinclair N, Endres M and Painter O 2019 Telecom-band quantum optics with ytterbium atoms and silicon nanophotonics *Phys. Rev. Appl.* **11** 034044
- [47] Menon S G, Singh K, Borregaard J and Bernien H 2020 Nanophotonic quantum network node with neutral atoms and an integrated telecom interface *New J. Phys.* **22** 073033
- [48] Kane B E 1998 A silicon-based nuclear spin quantum computer *Nature* **393** 133
- [49] Petit L 2020 Universal quantum logic in hot silicon qubits *Nature* **580** 355
- [50] He Y, Gorman S K, Keith D, Kranz L, Keizer J G and Simmons M Y 2019 A two-qubit gate between phosphorus donor electrons in silicon *Nature* **571** 371
- [51] Maqdzik M T, Laucht A, Hudson F E, Jakob A M, Johnson B C, Jamieson D N, Itoh K M, Dzurak A S and Morello A 2021 Conditional quantum operation of two exchange-coupled single-donor spin qubits in a MOS-compatible silicon device *Nat. Commun.* **12** 181
- [52] Pla J J, Tan K Y, Dehollain J P, Lim W H, Morton J J L, Zwanenburg F A, Jamieson D N, Dzurak A S and Morello A 2013 High-fidelity readout and control of a nuclear spin qubit in silicon *Nature* **496** 334
- [53] Pla J J, Tan K Y, Dehollain J P, Lim W H, Morton J J L, Jamieson D N, Dzurak A S and Morello A 2012 A single-atom electron spin qubit in silicon *Nature* **489** 541
- [54] Maqdzik M T et al 2021 Precision tomography of a three-qubit electron-nuclear quantum processor in silicon (arXiv: 2106.03082v2)

- [55] Muhonen J T *et al* 2014 Storing quantum information for 30 seconds in a nanoelectronic device *Nat. Nanotechnol.* **9** 986
- [56] Weber B, Tan Y H M, Mahapatra S, Watson T F, Ryu H, Rahman R, Hollenberg L C L, Klimeck G and Simmons M Y 2014 Spin blockade and exchange in Coulomb-confined silicon double quantum dots *Nat. Nanotechnol.* **9** 430
- [57] Keith D *et al* 2019 Single-shot spin readout in semiconductors near the shot-noise sensitivity limit *Phys. Rev. X* **9** 041003
- [58] Weber B, Hsueh Y-L, Watson T F, Li R, Hamilton A R, Hollenberg L C L, Rahman R and Simmons M Y 2018 Spin-orbit coupling in silicon for electrons bound to donors *NPJ Quantum Inf.* **4** 61
- [59] Steger M, Saeedi K, Thewalt M L W, Morton J J L, Riemann H, Abrosimov N V, Becker P and Pohl H-J 2012 Quantum information storage for over 180 s using donor spins in a ^{28}Si 'Semiconductor Vacuum' *Science* **336** 1280
- [60] Feher G and Gere E A 1959 Electron spin resonance experiments on donors in Silicon II: electron spin relaxation effects *Phys. Rev.* **114** 1245
- [61] Tyryshkin A M *et al* 2012 Electron spin coherence exceeding seconds in high-purity silicon *Nat. Mater.* **11** 143
- [62] Saeedi K, Simmons S, Salvail J Z, Dluhy P, Riemann H, Abrosimov N V, Becker P, Pohl H-J, Morton J J L and Thewalt M L W 2013 Room-temperature quantum bit storage exceeding 39 minutes using ionized donors in silicon-28 *Science* **342** 830
- [63] Wang Y, Tankasala A, Hollenberg L C L, Klimeck G, Simmons M Y and Rahman R 2016 Highly tunable exchange in donor qubits in silicon *NPJ Quantum Inf.* **2** 16008
- [64] Jakob A M *et al* 2020 Deterministic single ion implantation with 99.87% confidence for scalable donor-qubit arrays in silicon (arXiv:2009.02892)
- [65] Krauth F N *et al* 2021 Flopping-mode electric dipole spin resonance in phosphorus donor qubits in silicon (arXiv:2105.029061)
- [66] Tosi G, Mohiyaddin F A, Schmitt V, Tenberg S, Rahman R, Klimeck G and Morello A 2017 Silicon quantum processor with robust long-distance qubit couplings *Nat. Commun.* **8** 450
- [67] Hill C D, Peretz E, Hile S J, House M G, Fuechsle M, Rogge S, Simmons M Y and Hollenberg L C L 2015 A surface code quantum computer in silicon *Sci. Adv.* **1** e1500707
- [68] Doherty M W, Manson N B, Delaney P, Jelezko F, Wrachtrup J and Hollenberg L C L 2013 The nitrogen-vacancy colour centre in diamond *Phys. Rep.* **528** 1–45
- [69] Atatüre M, Englund D, Vamivakas N, Lee S-Y and Wrachtrup J 2018 Material platforms for spin-based photonic quantum technologies *Nat. Rev. Mater.* **3** 38–51
- [70] Rondin L, Tietjenne J P, Hingant T, Roch J F, Maletinsky P and Jacques V 2014 Magnetometry with nitrogen-vacancy defects in diamond *Rep. Prog. Phys.* **77** 056503
- [71] Barry J F, Schloss J M, Bauch E, Turner M J, Hart C A, Pham L M and Walsworth R L 2020 Sensitivity optimization for NV-diamond magnetometry *Rev. Mod. Phys.* **92** 015004
- [72] Schmitt S *et al* 2017 Submillihertz magnetic spectroscopy performed with a nanoscale quantum sensor *Science* **356** 832–7
- [73] Bucher D B, Glenn D R, Park H, Lukin M D and Walsworth R L 2020 Hyperpolarization-enhanced NMR spectroscopy with femtomole sensitivity using quantum defects in diamond *Phys. Rev. X* **10** 021053
- [74] Bradley C E, Randall J, Abobeih M, Berrevoets R, Degen M, Bakker M, Markham M, Twitche D and Taminiau T 2019 A ten-qubit solid-state spin register with quantum memory up to one minute *Phys. Rev. X* **9** 031045
- [75] Pompili M *et al* 2021 Realization of a multinode quantum network of remote solid-state qubits *Science* **372** 259–64
- [76] Zaitsev A M *et al* 2001 *Optical Properties of Diamond* (Berlin: Springer)
- [77] Bradac C, Gao W, Forneris J, Trusheim M E and Aharonovich I 2019 Quantum nanophotonics with group IV defects in diamond *Nat. Commun.* **10** 5625
- [78] Bhaskar M K *et al* 2020 Experimental demonstration of memory-enhanced quantum communication *Nature* **580** 60–4
- [79] Wan N H *et al* 2020 Large-scale integration of artificial atoms in hybrid photonic circuits *Nature* **583** 226–31
- [80] Lukin D M, Guidry M A and Vučković J 2020 Integrated quantum photonics with silicon carbide: challenges and prospects *PRX Quantum* **1** 020102
- [81] Anderson C P *et al* 2022 Five-second coherence of a single spin with single-shot readout in silicon carbide *Sci. Adv.* **8** eabm5912
- [82] Gottscholl A *et al* 2020 Initialization and read-out of intrinsic spin defects in a van der Waals crystal at room temperature *Nat. Mater.* **19** 540–5
- [83] Gottscholl A, Diez M, Soltamov V, Kasper C, Sperlich A, Kianinia M, Bradac C, Aharonovich I and Dyakonov V 2021 Room temperature coherent control of spin defects in hexagonal boron nitride *Sci. Adv.* **7** eabf3630
- [84] Mendelson N *et al* 2021 Coupling spin defects in a layered material to nanoscale plasmonic cavities *Adv. Mater.* **34** e2106046
- [85] Herbschleb E D *et al* 2019 Ultra-long coherence times amongst room-temperature solid-state spins *Nat. Commun.* **10** 3766
- [86] Toyli D M, Christle D J, Alkauskas A, Buckley B B, Van de Walle C G and Awschalom D D 2012 Measurement and control of single nitrogen-vacancy center spins above 600 K *Phys. Rev. X* **2** 031001
- [87] Sukachev D D, Sipahigil A, Nguyen C, Bhaskar M, Evans R, Jelezko F and Lukin M 2017 Silicon-vacancy spin qubit in diamond: a quantum memory exceeding 10 ms with single-shot state readout *Phys. Rev. Lett.* **119** 223602
- [88] Sipahigil A, Jahnke K, Rogers L, Teraji T, Isoya J, Zibrov A, Jelezko F and Lukin M 2014 Indistinguishable photons from separated silicon-vacancy centers in diamond *Phys. Rev. Lett.* **113** 113602
- [89] Chen D *et al* 2022 Quantum interference of resonance fluorescence from germanium-vacancy color centers in diamond *Nano Lett.* **22** 6306–12
- [90] Thiering G and Gali A 2018 *Ab initio* magneto-optical spectrum of group-IV vacancy color centers in diamond *Phys. Rev. X* **8** 021063
- [91] Trusheim M E *et al* 2020 Transform-limited photons from a coherent tin-vacancy spin in diamond *Phys. Rev. Lett.* **124** 023602
- [92] Debroux R *et al* 2021 Quantum control of the tin-vacancy spin qubit in diamond *Phys. Rev. X* **11** 041041
- [93] Görlitz J *et al* 2020 Spectroscopic investigations of negatively charged tin-vacancy centres in diamond *New J. Phys.* **22** 013048
- [94] Wang P, Taniguchi T, Miyamoto Y, Hatano M and Iwasaki T 2021 Low-temperature spectroscopic investigation of lead-vacancy centers in diamond fabricated by high-pressure and high-temperature treatment *ACS Photonics* **8** 2947–54
- [95] Christle D J *et al* 2017 Isolated spin qubits in SiC with a high-fidelity infrared spin-to-photon interface *Phys. Rev. X* **7** 021046
- [96] Jurgen von Bardeleben H, Cantin J-L, Gerstmann U, Schmidt W G and Biktagirov T 2021 Spin polarization, electron-phonon coupling, and zero-phonon line of the NV center in 3C-SiC *Nano Lett.* **21** 8119–25
- [97] Sörman E, Son N, Chen W, Kordina O, Hallin C and Janzén E 2000 Silicon vacancy related defect in 4H and 6H SiC *Phys. Rev. B* **61** 2613
- [98] Li Q *et al* 2022 Room-temperature coherent manipulation of single-spin qubits in silicon carbide with a high readout contrast *Nat'l Sci. Rev.* **9** nwab122

- [99] Wang J-F et al 2020 Coherent control of nitrogen–vacancy center spins in silicon carbide at room temperature *Phys. Rev. Lett.* **124** 223601
- [100] Wolfowicz G, Anderson C P, Diler B, Poluektov O G, Heremans F J and Awschalom D D 2020 Vanadium spin qubits as telecom quantum emitters in silicon carbide *Sci. Adv.* **6** eaaz1192
- [101] Falk A L, Buckley B B, Calusine G, Koehl W F, Dobrovitski V V, Politi A, Zorman C A, Feng P X-L and Awschalom D D 2013 Politype control of spin qubits in silicon carbide *Nat. Commun.* **4** 1819
- [102] Khazen K, von Bardeleben H J, Zargaleh S A, Cantin J L, Zhao M, Gao W, Biktagirov T and Gerstmann U 2019 High-resolution resonant excitation of NV centers in 6H–SiC: a matrix for quantum technology applications *Phys. Rev. B* **100** 205202
- [103] Gao X et al 2021 High-contrast plasmonic-enhanced shallow spin defects in hexagonal boron nitride for quantum sensing *Nano Lett.* **21** 7708–14
- [104] Qian C et al 2022 Unveiling the zero-phonon line of the boron vacancy center by cavity enhanced emission (arXiv:2202.10980)
- [105] Schröder T et al 2016 Quantum nanophotonics in diamond *J. Opt. Soc. Am. B* **33** B65–B83
- [106] Sangtawesin S et al 2019 Origins of diamond surface noise probed by correlating single-spin measurements with surface spectroscopy *Phys. Rev. X* **9** 031052
- [107] Yuan Z, Fitzpatrick M, Rodgers L V H, Sangtawesin S, Srinivasan S and de Leon N P 2020 Charge state dynamics and optically detected electron spin resonance contrast of shallow nitrogen–vacancy centers in diamond *Phys. Rev. Res.* **2** 033263
- [108] Wolfowicz G et al 2021 Quantum guidelines for solid-state spin defects *Nat. Rev. Mater.* **6** 906–25
- [109] Sohn Y I et al 2018 Controlling the coherence of a diamond spin qubit through its strain environment *Nat. Commun.* **9** 2012
- [110] Whiteley S J et al 2019 Spin–phonon interactions in silicon carbide addressed by Gaussian acoustics *Nat. Phys.* **15** 490–5
- [111] Maity S et al 2020 Coherent acoustic control of a single silicon vacancy spin in diamond *Nat. Commun.* **11** 193
- [112] MacQuarrie E R, Gosavi T A, Jungwirth N R, Bhawe S A and Fuchs G D 2013 Mechanical spin control of nitrogen–vacancy centers in diamond *Phys. Rev. Lett.* **111** 227602
- [113] Dietz J R et al Spin-acoustic control of silicon vacancies in 4H silicon carbide (arXiv:2205.15488)
- [114] Zhou H et al 2020 Quantum metrology with strongly interacting spin systems *Phys. Rev. X* **10** 031003
- [115] Kanai S et al 2022 Generalized scaling of spin qubit coherence in over 12,000 host materials *Proc. Natl Acad. Sci.* **119** e2121808119
- [116] Abdi M, Chou J-P, Gali A and Plenio M B 2018 Color centers in hexagonal boron nitride monolayers: a group theory and *ab initio* analysis *ACS Photonics* **5** 1967–76
- [117] Tsai J-Y, Pan J, Lin H, Bansil A and Yan Q 2022 Antisite defect qubits in monolayer transition metal dichalcogenides *Nat. Commun.* **13** 492
- [118] Loss D and DiVincenzo D P 1998 Quantum computation with quantum dots *Phys. Rev. A* **57** 120–6
- [119] Kormányos A, Zólyomi V, Drummond N D and Burkard G 2014 Spin–orbit coupling, quantum dots, and qubits in monolayer transition metal dichalcogenides *Phys. Rev. X* **4** 011034
- [120] Brotons-Gisbert M et al 2019 Coulomb blockade in an atomically thin dichalcogenide dot coupled to a tunable Fermi reservoir *Nat. Nanotechnol.* **14** 442–6
- [121] He Y-M et al 2015 Single quantum emitters in monolayer semiconductors *Nat. Nanotechnol.* **10** 497–502
- [122] Koperski M et al 2015 Single photon emitters in exfoliated WSe₂ structures *Nat. Nanotechnol.* **10** 503–6
- [123] Tran T T, Bray K, Ford M J, Toth M and Aharonovich I 2016 Quantum emission from hexagonal boron nitride monolayers *Nat. Nanotechnol.* **11** 37–41
- [124] Baek H et al 2020 Highly energy-tunable quantum light from moiré-trapped excitons *Sci. Adv.* **6** aba8526
- [125] Gottscholl A et al 2021 Spin defects in hBN as promising temperature, pressure and magnetic field quantum sensors *Nat. Commun.* **12** 4480
- [126] Dreyer C E, Alkauskas A, Lyons J L, Janotti A and Van de Walle C G 2018 First-principles calculations of point defects for quantum technologies *Annu. Rev. Mater. Res.* **48** 1–26
- [127] Kennes D M, Claassen M, Xian L, Georges A, Millis A J, Hone J, Dean C R, Basov D N, Pasupathy A N and Rubio A 2021 Moiré heterostructures as a condensed-matter quantum simulator *Nat. Phys.* **17** 155–63
- [128] Gjerding M N et al 2021 Recent progress of the computational 2D materials database (C2DB) *2D Mater.* **8** 044002
- [129] Michler P, Kiraz A, Becher C, Schoenfeld W V, Petroff P M, Zhang L, Hu E and Imamoglu A 2000 A quantum dot single-photon turnstile device *Science* **290** 2282–5
- [130] Benson O, Santori C, Pelton M and Yamamoto Y 2000 Regulated and entangled photons from a single quantum dot *Phys. Rev. Lett.* **84** 2513
- [131] Mar J D, Baumberg J J, Xu X L, Irvine A C and Williams D A 2017 Precise measurements of the dipole moment and polarizability of the neutral exciton and positive trion in a single quantum dot *Phys. Rev. B* **95** 201304(R)
- [132] Senellart P, Solomon G and White A 2017 High-performance semiconductor quantum-dot single-photon sources *Nat. Nanotechnol.* **12** 1026–39
- [133] Press D, Ladd T D, Zhang B and Yamamoto Y 2008 Complete quantum control of a single quantum dot spin using ultrafast optical pulses *Nature* **456** 218–21
- [134] Tomm N et al 2021 A bright and fast source of coherent single photons *Nat. Nanotechnol.* **16** 399–403
- [135] Gimeno-Segovia M, Rudolph T and Economou S E 2019 Deterministic generation of large-scale entangled photonic cluster state from interacting solid state emitters *Phys. Rev. Lett.* **123** 070501
- [136] Bopp F et al 2022 Quantum dot molecule devices with optical control of charge status and electronic control of coupling (arXiv:2205.10001)
- [137] Wang H et al 2019 Boson sampling with 20 input photons and a 60-mode interferometer in a 10¹⁴-dimensional Hilbert space *Phys. Rev. Lett.* **123** 250503
- [138] Zaporski L et al 2022 Ideal refocusing of an optically active spin qubit under strong hyperfine interactions (arXiv:2206.01223)
- [139] Schmidt O G (ed) 2007 *Lateral Alignment of Quantum Dots* (Berlin: Springer)
- [140] Prechtel J et al 2013 Frequency-stabilized source of single photons from a solid-state qubit *Phys. Rev. X* **3** 041006
- [141] Bayer M, Kuther A, Forchel A, Gorbunov A, Timofeev V B, Schäfer F, Reithmaier J P, Reinecke T L and Walck S N 1999 Electron and hole g-factors and exchange interaction from studies of the exciton fine structure in In_{0.60}Ga_{0.40}As quantum dots *Phys. Rev. Lett.* **82** 1748
- [142] Trotta R, Zallo E, Ortix C, Atkinson P, Plumhof J D, van den Brink J, Rastelli A and Schmidt O G 2012 Universal recovery of the energy-level degeneracy of bright excitons in InGaAs quantum dots without a structure symmetry *Phys. Rev. Lett.* **109** 147401

- [143] Manga Rao V S C and Hughes S 2007 Single quantum dot spontaneous emission in a finite-size photonic crystal waveguide: proposal for an efficient 'On Chip' single photon gun *Phys. Rev. Lett.* **99** 193901
- [144] Santori C, Fattal D, Vučković J, Solomon G S and Yamamoto Y 2002 Indistinguishable photons from a single-photon device *Nature* **419** 594–7
- [145] Gazzano O, Huber T, Loo V, Polyakov S, Flagg E B and Solomon G S 2018 Effects of resonant-laser excitation on the emission properties in a single quantum dot *Optica* **5** 354
- [146] Huber T, Predojević A, Föger D, Solomon G and Weihs G 2015 Optimal excitation conditions for indistinguishable photons from quantum dots *New J. Phys.* **17** 123025
- [147] Gol'tsman G N, Okunev O, Chulkova G, Lipatov A, Semenov A, Smirnov K, Voronov B, Dzardanov A, Williams C and Sobolewski R 2001 Picosecond superconducting single-photon optical detector *Appl. Phys. Lett.* **79** 705–7
- [148] Reddy D V, Nerem R R, Nam S W, Mirin R P and Verma V B 2020 Superconducting nanowire single-photon detectors with 98% system detection efficiency at 1550 nm *Optica* **7** 1649–53
- [149] Chang J et al 2021 Detecting telecom single photons with 99.5 – 2.07 + 0.5% system detection efficiency and high time resolution *APL Photonics* **6** 036114
- [150] Korzh B et al 2020 Demonstration of sub-3 ps temporal resolution with a superconducting nanowire single-photon detector *Nat. Photon.* **14** 250–5
- [151] Hochberg Y, Charaev I, Nam S W, Verma V, Colangelo M and Berggren K K 2019 Detecting sub-GeV dark matter with superconducting nanowires *Phys. Rev. Lett.* **123** 151802
- [152] Cahall C, Nicolich K L, Islam N T, Lafyatis G P, Miller A J, Gauthier D J and Kim J 2017 Multi-photon detection using a conventional superconducting nanowire single-photon detector *Optica* **4** 1534–5
- [153] Zhu D, Colangelo M, Chen C, Korzh B A, Wong F N C, Shaw M D and Berggren K K 2020 Resolving photon numbers using a superconducting nanowire with impedance-matching taper *Nano Lett.* **20** 3858–63
- [154] Ferrari S, Schuck C and Pernice W 2018 Waveguide-integrated superconducting nanowire single-photon detectors *Nanophotonics* **7** 1725–58
- [155] Holzman I and Ivry Y 2019 Superconducting nanowires for single-photon detection: progress, challenges, and opportunities *Adv. Quantum Technol.* **2** 1800058
- [156] Verma V B et al 2021 Single-photon detection in the mid-infrared up to 10 μm wavelength using tungsten silicide superconducting nanowire detectors *APL Photonics* **6** 056101
- [157] Allmaras J P, Kozorezov A G, Korzh B A, Berggren K K and Shaw M D 2019 Intrinsic timing jitter and latency in superconducting nanowire single-photon detectors *Phys. Rev. Appl.* **11** 034062
- [158] Steinhauer S, Gyger S and Zwiller V 2021 Progress on large-scale superconducting nanowire single-photon detectors *Appl. Phys. Lett.* **118** 100501
- [159] Cheng R, Wright J, Xing H G, Jena D and Tang H X 2020 Epitaxial niobium nitride superconducting nanowire single-photon detectors *Appl. Phys. Lett.* **117** 132601
- [160] Taylor G G, Morozov D V, Lennon C T, Barry P S, Sheagren C and Hadfield R H 2021 Infrared single-photon sensitivity in atomic layer deposited superconducting nanowires *Appl. Phys. Lett.* **118** 191106
- [161] Cherednichenko S, Acharya N, Novoselov E and Drakinskiy V 2021 Low kinetic inductance superconducting MgB_2 nanowires with a 130ps relaxation time for single-photon detection applications *Supercond. Sci. Technol.* **34** 044001
- [162] Lyatti M, Wolff M A, Gundareva I, Kruth M, Ferrari S, Dunin-Borkowski R E and Schuck C 2020 Energy-level quantization and single-photon control of phase slips in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ nanowires *Nat. Commun.* **11** 763