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Challenges in advancing our understanding of atomic-like quantum systems: Theory and experiment

Adam Gali, André Schleife,* Andreas J. Heinrich, Arne Laucht, Bruno Schuler, Chitraleema Chakraborty, Christopher P. Anderson, Corentin Déprez, Jeffrey McCallum, Lee C. Bassett, Mark Friesen, Michael E. Flatté, Peter Maurer, Susan N. Coppersmith, Tian Zhong, Vijaya Begum-Hudde, and Yuan Ping

Quantum information processing and quantum sensing is a central topic for researchers who are part of the Materials Research Society and the Quantum Staging Group is providing leadership and guidance in this context. We convened a workshop before the 2022 MRS Spring Meeting and covered four topics to explore challenges that need to be addressed to further promote and accelerate the development of materials with applications in quantum technologies. This article captures the discussions at this workshop and refers to the pertinent literature.

Introduction

The mission of the Quantum Staging Group (QSG) is to provide leadership and guidance for the Materials Research Society (MRS) in promotion of materials science for development of quantum information sciences and quantum sensing. The QSG provides advice and support to international leaders in the field of quantum information science and engineering on symposium and workshop development and other activities

Adam Gali, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; Department of Atomic Physics, Institute of Physics, Budapest University of Technology and Economics, Budapest, Hungary; gali.adam@wigner.hu
André Schleife, Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, USA; Materials Research Laboratory, University of Illinois at Urbana-Champaign, Urbana, USA; National Center for Supercomputing Applications, University of Illinois at Urbana-Champaign, Urbana, USA; schleife@illinois.edu
Andreas J. Heinrich, Center for Quantum Nanoscience, Institute for Basic Science, Seoul, South Korea; Department of Physics, Ewha Womans University, Seoul, South Korea; heinrich.andreas@qns.science
Arne Laucht, School of Electrical Engineering and Telecommunications, University of New South Wales, Sydney, NSW, Australia; Diraq Pty Ltd., Sydney, NSW, Australia; a.laucht@unsw.edu.au
Bruno Schuler, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; nanotech@surfaces Laboratory, Empa – Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland; Bruno.Schuler@empa.ch
Chitraleema Chakraborty, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; Department of Materials Science and Engineering, University of Delaware, Newark, USA; Department of Physics and Astronomy, University of Delaware, Newark, USA; cchakrab@udel.edu
Christopher P. Anderson, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, USA; E.L. Ginzton Laboratory, Stanford University, Stanford, USA; cpand@illinois.edu
Corentin Déprez, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; QuTech and Kavli Institute of Nanoscience, Delft University of Technology, GA, Delft, The Netherlands; C.C.Deprez@tudelft.nl
Jeffrey McCallum, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; School of Physics, University of Melbourne, Melbourne, VIC, Australia; jeffreym@unimelb.edu.au
Lee C. Bassett, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; Quantum Engineering Laboratory, Department of Electrical and Systems Engineering, University of Pennsylvania, Philadelphia, USA; lbassett@seas.upenn.edu
Mark Friesen, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; Department of Physics, University of Wisconsin–Madison, Madison, USA; friesen@physics.wisc.edu
Michael E. Flatté, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; Department of Physics and Astronomy, The University of Iowa, Iowa City, USA; Department of Applied Physics, Technische Universiteit Eindhoven, Eindhoven, The Netherlands; michael-flatte@uiowa.edu
Peter Maurer, Department of Atomic Physics, Institute of Physics, Budapest University of Technology and Economics, Budapest, Hungary; Pritzker School of Molecular Engineering, The University of Chicago, Chicago, USA; Center for Molecular Engineering and Materials Science Division, Argonne National Laboratory, Lemont, USA; pmaurer@uchicago.edu
Susan N. Coppersmith, HUN-REN Wigner Research Centre for Physics, Budapest, Hungary; Department of Atomic Physics, Institute of Physics, Budapest University of Technology and Economics, Budapest, Hungary; Department of Physics, University of Wisconsin–Madison, Madison, USA; School of Physics, University of New South Wales, Sydney, NSW, Australia; s.coppersmith@unsw.edu.au
Tian Zhong, Department of Atomic Physics, Institute of Physics, Budapest University of Technology and Economics, Budapest, Hungary; Pritzker School of Molecular Engineering, The University of Chicago, Chicago, USA; tzh@uchicago.edu
Vijaya Begum-Hudde, Department of Materials Science and Engineering, University of Illinois at Urbana-Champaign, Urbana, USA; vijayab@illinois.edu
Yuan Ping, Department of Atomic Physics, Institute of Physics, Budapest University of Technology and Economics, Budapest, Hungary; Department of Materials Science and Engineering, University of Wisconsin, Madison, USA; Department of Physics, University of California, Santa Cruz, USA; yuanping@ucsc.edu
*Corresponding author

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including development of publications that promote materials science for quantum technologies. It was formed in 2018 and has held a number of workshops and panel discussions linked to both the Spring and Fall meetings.

While the QSG provides support for quantum materials development across all platforms, including superconductors, ion traps, topological systems, and more, the particular focus of this workshop was solid-state atom-like systems, including impurity-based qubits in semiconductors and defects in two-dimensional (2D) and three-dimensional (3D) materials systems.

This article provides a summary of the QSG workshop titled "Challenges in Advancing Our Understanding of Atomic-Like Quantum Systems: Theory and Experiment," that was held at the 2022 MRS Spring Meeting in Hawaii. The workshop was held in conjunction with the meeting symposium QT07-Atomic and Molecular Quantum Systems and Defect Engineering for Quantum Technologies. The aim of the workshop was to bring together scientists with experimental and theoretical expertise in materials for quantum technologies to discuss key near-term challenges that need to be addressed to further promote and accelerate development of solid-state atom-like systems with applications in quantum technologies.

The main emphasis was on having both theoreticians and experimentalists come together to provide their perspectives on the opportunities and challenges associated with the underlying materials science needed to develop and control atom-like quantum entities across a range of platforms that form qubits, arrays of qubits, and long-range communication between qubit systems. As a result, the workshop promoted greater awareness of where key focus is required and what tools are needed to make real progress. There were four themes addressed in the workshop and summarized next: "Are there unifying perspectives on the relevant length scales for quantum systems? How to span from 1 nm to 1 micron?," "Decoherence at the interface: How can quantum nanoscience address materials challenges in quantum information technologies?," "Are there unifying perspectives on the role of electrical noise in atomic-like systems in materials?", and "What are the predictive challenges for atomic-like quantum systems?"

Unifying perspectives on spanning nanometer to micron-length scales

The first discussion was led by S. Coppersmith and focused on how materials questions that are critical to further progress keep evolving because of the tremendous advancements made in the development of qubits in Si/SiGe quantum dots. In 2022, three groups published reports of experiments achieving one- and two-qubit gate fidelities exceeding 99.5 %[1–3] as well as successful operation of a six-qubit processor.[4] With the successful demonstration of gate fidelities high enough to be compatible with the requirements of the current generation of quantum error correction protocols, the question of how to achieve extremely high yields of devices with this level of gate performance becomes critical. In the context of the entire workshop, this session highlighted recent progress on materials issues that are enabling substantial improvements in yields of devices that host qubits with high-fidelity gates. Both talks in the session highlighted how new key questions emerge as the field progresses and figuring out how to scale up to large numbers of qubits becomes a pressing concern.

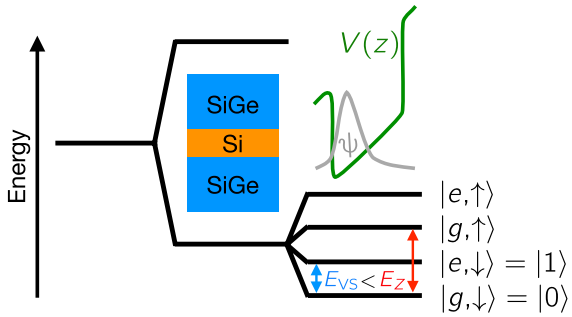


Figure 1. Valley splitting in a SiGe/Si/SiGe quantum well. Silicon conduction-band valleys are sixfold orbitally degenerate in bulk. Applying strain in a SiGe/Si/SiGe quantum well reduces the low-lying orbital degeneracy to twofold. Quantum-well confinement (green), combined with Zeeman splitting E_Z, fully lifts the ground-state degeneracy to give the valley splitting E_{VS}. Here, $|g\rangle$ and $|e\rangle$ refer to ground and excited valleys. However, details of the quantum-well interface could result in a low valley splitting ($E_{VS} < E_Z$), and an undesired level ordering, in which the ground-state manifold is not spin-like.

The work in References 1–4 was performed using quantum dots hosted in silicon/silicon-germanium (Si/SiGe) heterostructures, and M. Friesen's talk in the session focused on this materials system and on new understanding of how to fabricate quantum dots in which the lowest-lying excited states are reliably found to be spin states, rather than conduction-band valley states. The presentation began by introducing quantum dot (QD) spin qubits as systems that have the advantage of long-lived spin states, all-electrical control, and compatibility with existing classical electronics due to the choice of materials they are embedded in and made of. Widely employed GaAs QDs have the drawback of predominant nuclear spins, and thus, in recent years, the focus has shifted to Si and Ge materials.[5] The state-of-the-art one- and two-qubit gates show fidelities of >99.9[6] and 99.5%.

A major challenge in Si arises due to the sixfold orbital degeneracy of the lowest conduction band.[7] By applying biaxial strain, the fourfold *xy* and the twofold *z* valleys split by about 0.2 eV (**Figure** 1). A further valley splitting of <0.2 meV, between the low-lying $+z$ and $-z$ valley eigenstates, can be achieved by strongly confining the quantum dot within the quantum well.[8] However, the different spin states may be Zeeman split by about 0.2 meV, leaving the valley splitting as the defining energy scale for the qubits, rather than the spin splitting.

Although valley degeneracy can be lifted by means of a strong quantum-well confinement potential, the possibility of low-lying valley excited states arises because the valley minima occur at **k** vectors far from the zone center. In particular, small valley splittings can occur when there are no features in the confinement potential with large enough **k** vectors to couple the valleys. This problem is challenging to address theoretically because it is inherently multiscale–atomic-scale details of the quantum-well interface as well as the details of germanium concentration fluctuations in consecutive atomic layers play a critical role in determining the valley splitting. This understanding of the physics and the origins of the problem provides concrete avenues for developing heterostructure designs and growth processes that result in consistently high valley splittings.

The main challenge for achieving reproducibility of the valley splittings across a quantum dot device arises from unavoidable atomic-scale disorder. The dominant types of disorder in modern Si/SiGe quantum wells include (1) single-atom steps at interfaces, which can suppress valley splitting significantly (around 80%);[9–11] (2) Ge concentration fluctuations in the SiGe portion of the device, as Ge in Si forms a random alloy, resulting in a wide range of splitting between 0 and 250 μeV;[12] and (3) diffuse interfaces that are not perfectly sharp but spread out over several atomic layers, which can significantly suppress the splitting.[13] In many cases, valley splittings must be at least as large as the thermal energy scale in a dilution refrigerator ($k_BT \approx 10$–20 μeV), to enable high-fidelity initialization and readout. This is particularly true for qubit encodings known as Loss–DiVincenzo qubits[14] (i.e., a single-electron spin in a quantum dot). In other spin qubit encodings such as singlet-triplet qubits[15] (i.e., two electrons in a double quantum dot), quantum dot hybrid qubits[16] (i.e., three electrons in a double quantum dot), or exchange-only qubits[17] (i.e., three electrons in a triple quantum dot), large valley splittings are not necessarily a prerequisite for forming a well-defined qubit. However, good control and reproducibility of the valley splitting and other device parameters are always required for scaling up to many qubits.[18] The wide range of observed valley splittings, which vary significantly from dot to dot even within a single wafer, therefore poses a significant challenge for these systems. Although this variability can be mitigated slightly through carefully engineered heterostructures and gate control, it cannot be completely eliminated.

Deterministic solutions to the problem of valley splitting variability have recently been explored. The most promising of these is an approach known as the "Wiggle Well,"[19] wherein the *z* valleys are coupled by introducing specially chosen Ge concentration oscillations in the interior of the quantum well. This method has the potential of coupling valleys in the same Brillouin zone (short-period Wiggle Well) or in neighboring Brillouin zones (long-period Wiggle Well). The former structure will be challenging to grow in the laboratory, but it holds the promise of providing large, reproducible valley splittings 1.2 meV, which would certainly enable good quality spin qubits. To date, however, only the long-period Wiggle Well has been grown and measured.[19] Simulations confirm that the valley splitting enhancements provided by this structure are quite weak, and are overwhelmed by random-alloy disorder fluctuations of SiGe within the quantum well.

An alternative, nondeterministic approach to suppressing the variability of the valley splitting is to introduce uniform Ge into the quantum well.[13] In this case, natural Ge concentration fluctuations over the extent of a quantum dot enhance valley splitting, on average, by approximately mimicking the effect of a short-period wiggle well. A side effect of this statistical enhancement is the simultaneous increase of the variability of the valley splitting as a function of lateral position, due to increased overlap of the electron with the random alloy. To overcome this variability, simulations show that the ability to electrostatically control the position of the quantum dot over about 20 nm provides a mechanism for boosting the valley splitting to acceptable levels for the vast majority of devices on a given chip.[20]

In conclusion, Friesen emphasized the urgent need to develop atomic-scale growth and simulation techniques, to provide the level of control and uniformity needed for large-scale qubit implementations.

In the second talk in this session, C. Déprez presented recent results on a different materials system, holes in Ge/SiGe heterostructures, and discussed challenges in the scaling up of these hole spin qubits in germanium (see **Figure** 2a). Quantum dots with Ge holes have some advantages over Si electrons, because this materials system has no low-lying valley states, and it has strong spin–orbit coupling that obviates the need to fabricate lithographically patterned micromagnets in order to implement gate operations. Déprez presented state-of-the-art results in the context of the fabrication and the operation of a four-qubit processor[21] and of a sixteen-quantum dot device with shared gates.[22] The talk discussed physical effects that can limit the operation of large spin-qubit processors in Ge, such as variations in the hole *g*-factor caused by its strong dependence on the local electric field, both intrinsic and externally applied. While the applied external field used to manipulate the qubits is largest, electric fields from defects also affect qubit frequencies and need to be accounted for. The impact of nonuniformities of the confining potential on the operation of larger spin qubit devices was also discussed, and Déprez showed that they can be mitigated substantially by appropriate protocols for ramping gate voltages.

Semiconductors such as GaAs, Si/SiGe heterostructures, and SiMOS were employed for electron spin qubits and now SiMOS and Ge/SiGe are used for hosting hole spin qubits. Ge/SiGe heterostructures are particularly interesting for quantum information applications.[23] They were reported to host a 2D hole gas having a high mobility and they have a low charge noise level.[24–27] Further, holes in planar germanium have low effective mass, strong spin–orbit coupling, and an anisotropic and tunable *g*-factor. In the last few years, operations of all-electrically controlled single spin qubits[28,29] and

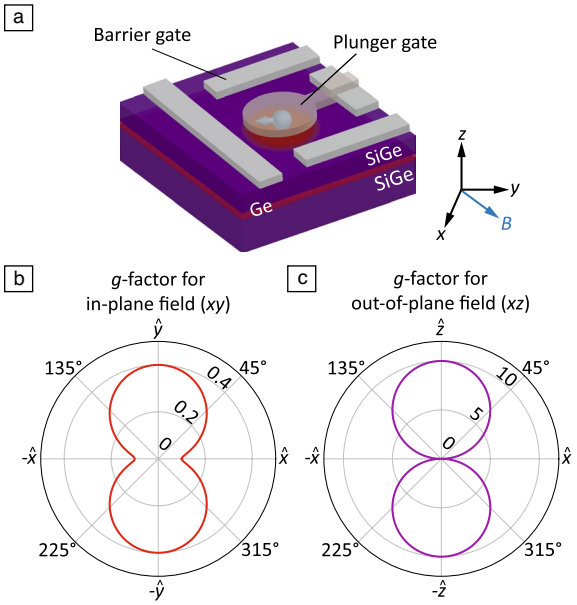


Figure 2. *g*-factor anisotropy in germanium spin qubit devices. (a) Schematic of a germanium hole quantum dot hosting a single spin qubit. Holes are accumulated beneath the plunger gate and confined using the barrier gates. An external magnetic field B is applied to split the spin states and to define a qubit. (b, c) Examples of *g*-factor evolution with the magnetic field orientation. The field orientation is described in the basis (*xyz*) formed by the principal axes of the *g*-tensor. The effective *g*-factor is then given by $\sqrt{(g_x B_x)^2 + (g_y B_y)^2 + (g_z B_z)^2}/|B|$, where (g_x, g_y, g_z) are the *g*-factor values along the principal axes.[36] The graphs are generated using typical values for (g_x, g_y, g_z) of (0.1, 0.4, 10).

of singlet-triplet qubits[30–32] based on hole quantum dots in planar germanium have been shown. They have been extended to implement a four-qubit quantum circuit[21] and a quantum simulation[32] using a 2D array of germanium quantum dots.

The *g*-factor in germanium depends strongly on the local electric field[28,30,31,33] and is highly anisotropic with $|g_\parallel| \approx 0.1 - 0.4$ and $g_\perp \approx 5 - 15$,[21,24,25,28–31,33–35] as illustrated in Figure 2b–c. These properties affect the operation of hole spin qubits and could challenge their scaling up. The strong dependence of the *g*-factor on the electric field enhances the effect of charge noise: it converts fluctuations of the electric environment into fluctuations of the Zeeman splitting, which affects coherence of single spin qubits. This dependence also leads to significant variations of the *g*-factor values inside the same device. It can even change sign for in-plane magnetic field configurations.[31] This spoils the initialization of singlet-triplet $S - T_0$ qubits as it leads to leakages to the T^- states. The *g*-factor variability also implies that the dynamics of singlet-triplet qubits within the same chip can be quite different. It complicates the practical operation of devices composed of multiple singlet-triplet qubits. For single spin qubits, large variations of the *g*-factors within a single device could also lead to an overhead in the control electronics used to drive the qubits.

Furthermore, the anisotropy of the *g*-factor also impacts the coherence of hole qubits via the hyperfine interaction. In 2D hole systems, the hyperfine interaction is also highly anisotropic such that the hole spin mostly couples to the *z*-components (out-of-plane) of the nuclear spins.[37,38] Thus, spin qubits in natural germanium are usually operated with an in-plane magnetic field. In practice, the external field is always slightly misaligned with the in-plane direction. Hence, the strong *g*-factor anisotropy magnifies the effects of this misalignment and amplifies the effect of hyperfine couplings affecting qubit coherence.[33,39] Note that the hyperfine interaction could be suppressed by using isotopically purified germanium, which would lead to an increase of the qubit coherence.[39,40]

To reduce dephasing and errors due to *g*-factor properties, solutions need to be found that allow to minimize the local variations of the electric field and the inhomogeneities in the confining potentials. One approach to achieve this is to increase the material uniformity, for example, by lowering the number of defects in the heterostructures or in the oxide layers, and in improving nanofabrication processes, for instance by using techniques that allow to fabricate gates having more uniform and more regular shapes. This would reduce the variations in the confining electric fields between quantum dots and thus in the *g*-factor values. Likewise, we could envision to adapt the design of quantum dots and engineer the confining potentials in order to make the *g*-factor less sensitive to electric field fluctuations.

Alternatively, one could take advantage of the *g*-factor properties to facilitate operation of hole spin qubit processors. Theoretical investigations[41,42] and experiments on different hole spin qubit platforms[33,43,44] suggest that these properties can lead to the emergence of "sweet spots" (i.e., operation points where individual qubits are insensitive to charge noise to the first order or, at least, less sensitive to it). These sweet spots can be found by changing the electric field strength and the magnetic field orientation. Finding sweet spots for multiple qubits, ways to induce them, or strategies to harvest the presence of individual sweet spots for multi-qubit operations would be of great interest as charge noise is likely the limiting noise source in germanium spin qubits.[28,33,39]

More generally, the scaling up of spin qubits requires the need to develop large 2D arrays of quantum dots, controlled using shared gates.[45–47] Current architectures where each gate is controlled via its exclusive voltage source are not scalable.[48] Despite significant improvements in the material growth and device fabrication, the control of 2D arrays with shared gate control is currently challenging due to local potential fluctuations.[22] It is unclear whether improvements in fabrication alone could lead to a sufficient degree of potential uniformity.

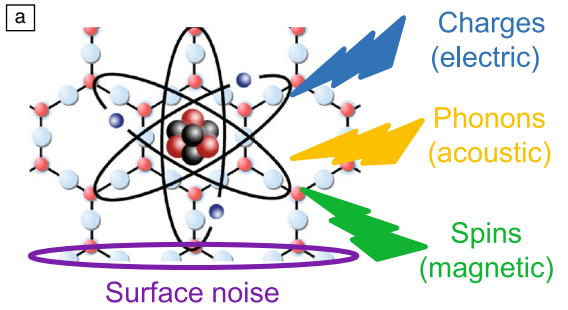
Solutions to compensate potential inhomogeneities after fabrication are, thus, required. One solution is to harvest hysteretic shifts in the potential landscape that appear after application of gate voltages. Tailored sequences of stress voltages can be used to tune the pinchoff voltages of individual gates and thereby the potential landscape beneath it.[49] Such

a method has been used to reduce the spread in the pinchoff voltages of gates inside a 1D quantum dot array in Si/SiGe by one order of magnitude, notably lowering the potential variations. Similar procedures can help in finding and tuning the operation points of quantum dot arrays and spin qubit devices.[50] They could also enable the control of devices with shared gates. The physical origin of these hysteretic shifts remains unclear and some studies suggest that they originate from charge traps at the heterostructure interfaces that are (un) filled by applying large gate voltages.[51–56] An understanding of the underlying mechanisms is needed and would help in designing heterostructures that allow these hysteretic effects to be further leveraged and new functionalities to be gained.

Nanoscale materials challenges in quantum information technologies

The second discussion topic of the workshop was led and introduced by A. Heinrich, and he raised the challenge of decoherence at the surface or at interfaces. The first contribution in this session focused on electron spins in semiconductors or insulators, which can be considered as solid-state analogs of ions and atoms in a vacuum chamber. As an automatic advantage, they are held in place by the crystal, making this aspect of the research much easier than ion and atom traps. However, one has to pay the price of additional decoherence sources stemming from interactions with the solid-state environment, rendering this a materials science research problem. When the spin qubits in insulators or semiconductors are brought close to the surface of the material or interfaces between materials, additional decoherence sources are typically found. On the flip-side, quantum-coherent control of spins on surfaces can be studied and achieved by combining electron spin resonance (ESR) spectroscopy and scanning tunneling microscopy (STM).[57] One area of active research is to find a spin system that can be attached to an STM tip and is ESR active. Such a system would vastly improve the spatial resolution of an NV center-based approach. On the other hand, it would require ultrahigh vacuum conditions and would most likely require low temperatures. In total, defects in solids can act as artificial trapped atoms, but their material environment causes electrical, magnetic, and acoustic noise that degrades performance. However, this noise is understood and can be mitigated by engineering the environment or tuning the qubit's properties.

In particular, C. Anderson discussed defects in wide-bandgap crystals, such as the negatively charged nitrogen-vacancy (NV) center in diamond, that can trap electrons and can be used to form a robust qubit. These electrons have both spin and optical structure, making them resemble trapped atoms in a solid-state environment (**Figure** 3a). Besides the NV center, there are also other emerging defects in diamond, and a wide range of qubit candidates in other materials.[58] This includes silicon carbide (SiC), silicon (Si), various 2D materials, oxides, and



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Problem	Materials solution
Spectral diffusion Charge instability	Symmetry Doping control Crystal purity
Short qubit lifetime Degraded optical properties	Temperature Stiffness/strain Size/dimensionality
Spin decoherence	Isotope control Hamiltonian engineering Crystal purity
Spin/optical/charge degradation	Surface termination Dangling bond passivation Reduced damage

Figure 3. Noise sources and materials solutions for atom-like defects. (a) Defects in solid-state materials are analogous to trapped atoms hosted in a crystal lattice, shown schematically. When integrated into the solid state, the dominant decoherence mechanisms are fluctuating charges (electrical noise, blue), coupling to phonons (acoustic noise, orange), and uncontrolled spins in the crystal, either nuclear or electronic (magnetic noise, green). Interfacial states, charge traps, and dangling bonds all are examples of types of surface noise (purple) that limit many applications in nanostructured devices or for quantum sensing. (b) These sources of noise cause problems for utilizing of these defects as qubits (left), but have materials engineering solutions to help mitigation (right). Text is color coded for the noise source in (a).

garnets such as yttrium orthovanadate (YVO) and yttrium aluminum garnet (YAG). The quantum states of these defects in the lattice are affected by phonons in the crystal (acoustic noise), other electron or nuclear spins (magnetic noise), and fluctuating charges (electric noise). These sources of noise also have additional considerations as the qubit approaches surfaces, which is relevant for quantum sensing applications or in nanofabricated devices. At the surface, dangling bonds and defects can be a source of additional unpaired electron spins, charge traps, or could even modify the phonon density of states.

Therefore, while the promise of these types of qubits is great, it is imperative that we understand and engineer these various noise sources. More generally, we also need to link the exact structure and Hamiltonian of the qubit to the susceptibility to this noise and the resulting coherence.[59] For

example, the point group symmetry of the defect in the host crystal determines the nature of the coupling to electric fields, as inversion symmetric defect sites are insensitive to electric fields to first order. For a highly coherent defect, it is important to eliminate any low-energy transitions that can be easily excited by excitations that do not couple to spin, such as phonons, for any operation above milli-Kelvin temperatures. Often, there are also tradeoffs where protecting from magnetic noise increases susceptibility to electrical noise in a qubit's Hamiltonian. Therefore, the exact materials requirements, such as reducing charge noise, isotope engineering, material stiffness, and more will depend on this interplay and the details of the quantum state. The highly interrelated nature of the materials properties, defect properties, and defect functionality for quantum computation, sensing, and communication[60] makes this a rich area for study.

For example, it is reasonable to identify preferred hosts for qubits based on the density, correlation time, and typical hopping distance of trapped charges, as these quantities will determine the spectral function of electric field noise that directly affects T_1 and T_2 that originates from impurities or defects. Temperature dependence will enter, for example, through the time scale for trapped charge hopping. Additional contributions to this spectral function would come from phonons, through direct, Raman, or Orbach processes that arise from the absorption, emission, or scattering of phonons in the crystal via the spin–orbit interaction. These relaxations are temperature-dependent and thus in turn decide the working temperature regime of the qubit, which can be dependent on the Debye temperature of the host crystal. Materials with high Debye temperature are therefore preferred for qubit operation at elevated temperatures, especially for quantum sensing. Phonons can also impact the optical structure, broadening lines, and reducing radiative efficiency. This is important when narrow wavelength, coherent photons are desired to mediate quantum entanglement between spins at a distance. Here, the common solution is simply to operate in appropriate cryogenic environments, where consideration needs to be made for the scalability of the necessary cryostat technology. Other possibilities for reducing this noise are creating a phononic bandgap, engineering the size or dimensionality of the crystal, or tuning the qubit's susceptibility to phonons via strain engineering.[61]

Magnetic noise is also substantially influenced by the crystal's chemistry and formation energies for defects. This noise is induced by the nuclear spins and unpaired electron spins residing in the host crystal, as the magnetic moments of these can fluctuate. These fluctuations cause uncontrolled noise that leads to loss of coherence. The dominant source of intrinsic magnetic field noise in a crystal without defects is often the nuclear spin of isotopes of a material, and their effects can be readily simulated by employing the fully quantum mechanical cluster correlation expansion (CCE) approach,[62] aiding in understanding decoherence and accurately predicting coherence times. This technique has even been used to screen more than 12,000 host materials for spin qubits in terms of coherence, breaking down contributions element-by-element, thus creating a new periodic table for spin coherence.[63] For systems with magnetically tunable optical lines, such noise can also impact the coherence of the emitted photons. Growth processes often influence the occurrence of unpaired electron spins; for example, in diamond doped with NV's, there is commonly a substantial presence of substitutional nitrogen that provides a spin bath decohering the NV.

In terms of engineering the host crystal to suppress magnetic noise, one can change the dimensionality using 2D systems,[64] or isotopically purify the material.[65] On the other hand, static and dynamic Hamiltonian engineering is extremely powerful for increasing coherence. For static protection, one needs a coupling between two subsystems (e.g., electrons and nuclear spins). This coupling hybridizes levels and causes anticrossing, which can be, to first order, insensitive to perturbations. For example, by tuning the magnetic field of certain hyperfine coupled systems, the qubit can be operated at a "clock" transition with a zero-first-order susceptibility to magnetic noise, and therefore increased coherence. For dynamic protection, one varies the number and type of quantum control pulses and drives, which is more universal and easy to implement in well-controlled systems. Such decoupling has been used to create the longest electron spin coherence in a solid with thousands of pulses.[66] In addition, if two-level systems are driven strongly, new dressed states are created, which are protected from noise. These two techniques can also be combined, creating a universally protected, highly coherent spin subspace.[67] From a materials science perspective, the coherence of these qubits can also be used as a probe of the nanoscale dynamics of spin, charge, and sound. In addition, using the previously mentioned techniques to extend coherences from elimination of one source of noise can often reveal new, underlying physical mechanisms in the material. Finally, the stated opportunities for coherence protection arise from exact details of crystal and defect site symmetry, hyperfine coupling, and the ability to grow custom materials with variable spin density and dimensionality, for example.

For magnetic noise at interfaces, proximity is key. At about 20 nm or closer, spin coherence becomes dominated by surface effects.[68] However, the noise-causing spins at the interface can be driven, creating an analog of motional narrowing that suppresses decoherence.[69] This extended coherence near surfaces is critical for nanoscale NMR applications.

Finally, electrical and charge noise limits these systems. In particular, the orbitals of defect qubits can have large electric dipole moments that make them susceptible to fluctuating charge traps or noisy surfaces in the solid state. This noise reduces the photon coherence, drastically lowering the rate and fidelity of entanglement generation through photon interference. In addition, uncontrolled charge environments can cause blinking and ionization, which affect most quantum applications. This noise can be understood and engineered by controlling the Fermi level or charge trap density in a material.[70] In this case, reducing the impurity concentration in materials is

key to reducing noise. For applications that require photonic integration of spins such as the creation of a quantum Internet,[71] electrical noise from surfaces is the number one hurdle toward scaling.

One technique unique to semiconductor systems is the use of a depletion region to remove fluctuating charges in the local environment of the defect. This has been used to improve both optical[72] and spin coherence times.[73] In addition to this depletion engineering, defects can be used that have a natural inversion symmetry of their structure in the lattice, making them resistant to electrical noise, displaying only a second-order susceptibility.[74,75] At surfaces, passivating charge traps can also stabilize lines.[76] However, for most systems, surfaces are still a problem. They cause band bending and unwanted charge instability,[77,78] or broaden optical transitions.[79] This surface noise can be understood theoretically,[80] and even reduced by dielectric passivation of surfaces for increased spin coherence.[81]

Although surfaces cause problems for many bulk spin qubits, they offer the unique capability of creating qubits by controlled absorption of atoms with partially filled electron shells or magnetic molecules.[57] This capability is enabled by embracing the need to control the surface of materials to the same level as the bulk, which generally requires the use of ultrahigh vacuum and surface science preparation techniques. Scanning tunneling microscopy is a powerful tool for such studies because it offers imaging of surfaces with atomic-scale spatial resolution in addition to being able to position atoms at will with atomic-scale precision and perform high-resolution tunneling spectroscopy *in situ*. Quantum-coherent control of spins on surfaces requires the incorporation of continuous-wave electron spin resonance,[82] which enabled a thousandfold increase in energy resolution over tunneling spectroscopy. Coherent control of atomic[83] and molecular spin qubits[84] was recently demonstrated through pulsed spin resonance. Although spins on surfaces offer the unique ability of atomic-scale control of nearest-neighbor interaction and artificially created nanostructures, they currently lack the long quantum coherence times that make defects in insulators so attractive.[84] Very recently, a technique was introduced to simultaneously control multiple spin qubits on a surface, which paves the way to a new electron spin qubit platform.[85] Further improvements in this very young research field will certainly require major advances in the materials science of surfaces.

Although there are many factors to consider, as a rule of thumb, single-defect qubit systems desire impurity levels below 10^{14} cm^{-3}, high purity single-crystal growth, and spin-bearing isotope fraction in the 0.1–1% range. ESR spectroscopy, DLTS, SIMs, and optical spectroscopy are all useful techniques for characterization. In addition, localized, deterministic defect creation is challenging, requiring advances in particle bombardment, atom-scale measurement, understanding of defect annealing kinetics, and first-principles calculations, for example. Finally, fabrication of devices and structures requires gentle, smooth, nondamaging etches with well-defined surface termination—providing a wealth of opportunities for materials scientists and engineers to contribute. Many of the major challenges for defect-based qubits can be mitigated by materials solutions (Figure 3b).

The second contribution in this session highlighted the possibilities and challenges stemming from the use of rare earth spins, which have quite unique and promising optical properties. Although RE qubits show excellent promise as communication and memory qubits for quantum networks, the significant gap in the optical and spin coherence times between bulk crystals and nanoscale devices poses a serious challenge that needs to be addressed. T. Zhong reported on engineering coherence of rare earth qubits in oxide host crystals, as a new platform based on epitaxial oxide thin films. Rare earth (RE) ions are well-known color centers when doped in solid-state host matrices, and have emerged in the last decade as a leading platform for optical quantum memories[86] that can simultaneously offer long spin coherence and narrow optical emission. Specifically, the 4 f–4 f intra-shell transitions in trivalent RE ions are effectively shielded by outer shells, giving rise to long optical coherence of up to milliseconds and spin coherence up to several hours. The engineering of RE-doped materials can build upon a rich history of RE spectroscopy.

In his discussion, Zhong focused on Kramers ions as spin qubits, where unpaired 4f electrons contribute to an effective spin 1/2 electronic state. For Kramers ions, the effective spin Hamiltonian includes contributions from spin–orbit coupling, crystal field interactions, Zeeman splitting, and hyperfine terms if both electronic and nuclear spins are present. The latter three terms can be engineered toward certain target metrics by host crystal selection, lattice site symmetry, external fields, and isotopic enrichment.

For quantum information science applications, the natural targets are optical quantum memory[86,88] and quantum repeaters, owing to the outstanding optical and spin coherence times demonstrated by rare earth dopants. This is further justified by a few particular rare earth ions such as erbium (Er) whose optical emission falls within the technologically relevant telecommunication band.[89] For example, as a primary candidate for long-distance quantum repeater networks, Er's telecom C-band (1530–1550 nm) emission is compatible with existing fiber-optic telecom infrastructure, which eliminates the need for any frequency conversion. For quantum repeater applications, there is a set of basic requirements,[90,91] two of which are as follows: First, long-distance repeater networks would require a spin coherence time of >1 ms to match with the 100 km inter-repeater distance considered appropriate for long-haul entanglement distribution. Second, indistinguishable photon emission from Er dopants is necessary for fusing entanglement between distant repeater nodes. Currently, both of these requirements are unmet, at least not simultaneously. The relatively short coherence time of Er

in prior work is mainly due to its large magnetic moments that make it susceptible to noise in the host matrix. The poor indistinguishability of Er photon emission is mainly due to the long radiative lifetime arising from the weak optical-dipole moment, and is exacerbated by optical spectral diffusion, especially when the dopants are positioned near defect-rich surfaces.

While there is a large body of work on achieving exceptionally long optical and spin coherence times of rare earth ions in bulk single crystals, there are only a few successful demonstrations to date on isolating individual rare earth ions with coherent control of the spin qubits. It is evident from recent work that there is a substantial discrepancy between the ensemble averaged optical/spin T_2 in bulk crystals versus their coherence as single qubits embedded in nanoscale photonic cavities.[92,93] The latter configuration is crucial to enhance the otherwise weak photoluminescence of rare earth emitters so that they can be read out efficiently.[94] However, embedding the rare earth emitters in photonic cavities also imposes significant excess decoherence due to proximity to etched surfaces. These challenges have impeded the development of high-quality telecom qubits for quantum repeater networks. It is thus important to develop an understanding of the different decoherence mechanisms impacting rare earth dopants in macroscopic bulk versus near surfaces of the host crystals.

One avenue to systematically study this is coherence spectroscopy on rare earth ions embedded in single-crystal thin films,[95,96] where the ions are intrinsically exposed to noise on the surfaces. By carefully selecting the local symmetry sites of rare earth occupancy, the coherence characteristics of the rare earth dopants can be used to reconstruct a comprehensive picture of the nanoscopic environment surrounding each individual dopant, including their interactions with structural defects, chemical impurities, and noise on the surface.

Zhong then discussed their study of Er in epitaxially grown Y_2O_3 thin films as a material test bed to elucidate decoherence mechanisms that are unique to this rare earth platform. Y_2O_3 is already shown to be an excellent host for Er ions, achieving sub-kilohertz optical linewidth[97] and millisecond long spin coherence.[98] Y_2O_3 can also be grown epitaxially on silicon substrates using molecular beam epitaxy[96] or other advanced thin-film deposition techniques. To perform both optical and spin coherence spectroscopy, they used the μ-electron spin resonance (ESR) technique and fiber optical cavities to measure spin and optical T_2 times of the Er qubits. In Y_2O_3, two symmetry sites exist, one with C_2 and the other with C_{3i} point group symmetries. The combination of both allows them to dissect the magnetic and electrical noises in the system,[87,99] as illustrated in **Figure** 4. Using dynamical decoupling techniques, we also reveal different time scales of the noise sources in the thin-film matrix.

This reveals sources of noise for rare earth qubits at the nanoscale, which in turn informs suitable strategies to suppress noise, in order to mitigate decoherence. First, growth

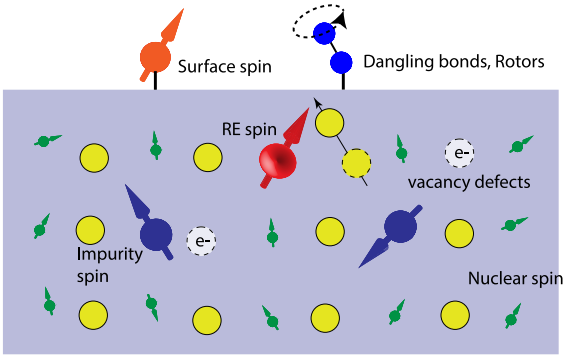

Figure 4. Magnetic and electrical noise sources in rare earth (RE) ion doped crystals and qubit devices. Magnetic TLS include paramagnetic impurities, surface spins, defects, RE spins in different lattice sites.[87] Electrical TLS includes dangling bonds and charged vacancy defects. TLS, two-level system.

of host materials with exceptionally high chemical purity and structural quality remains a priority. Part of this also involves using dopant source materials with ultrahigh purity such that paramagnetic impurities are minimized. This is particularly important for yttrium-based oxides in which, due to the long-standing difficulty in separating yttrium from other rare earth elements, background traces of other Kramers rare earth dopants introduce significant noise. Second, given the strong evidence of surface noise, treatment of the material surface such as passivation, desorption of surface spins (e.g., by vacuum annealing), and charge depletion will help to minimize decoherence. Third, exploiting site symmetry around the rare earth dopants could protect the relevant spin or optical transitions against decoherence. For instance, sites with centro-inversion symmetry quench the permanent electric dipole moment of the optical transition, which alleviates the optical spectral diffusion caused by electric noise. Fourth, for nuclear isotopes of rare earth ions with hybridized electro-nuclear spin transitions,[100] zero first-order Zeeman levels (ZEFOZ) are often present, which are addressed with less sensitivity to magnetic noise, leading to long spin coherence. Finally, there are methods to dynamically decouple rare earth ions from either electric or magnetic noise if the relevant transitions are driven out of equilibrium to generate new dressed noise tolerant eigenstates. In general, many of these approaches have been explored with reasonable success in NV, SiV, and SiC defect systems. Their extension to rare earth systems is expected to be effective as well.

Electrical noise in atomic-like systems in materials

The third discussion topic was chaired by M. Flatté who introduced how sensitivity to electric fields can be a problem for long spin coherence times, but can also be used for sensing (e.g., in nitrogen-vacancy [NV] centers). Spin centers in solids can have remarkably long room-temperature spin coherence times[101–103] and can be responsive to external fields, such

as electric fields,[104] that correspond to energy shifts of the spin center eigenstates that are miniscule compared to thermal energies. How can this be and still be consistent with thermodynamics?

There are several elements that conspire to allow the coherence times of these spins to be so long at room temperature.[105] First, the typical orbital and exchange energies (~1 eV for NV in diamond) are much larger than the thermal energies at room temperature (~1/40 eV), so all of the charge dynamics involving transitions between energy states is quenched for the spin center. Exceptions, for example, for the NV transitioning to an NV^0 state, are very destructive to quantum sensing applications.[78] Similarly, any other accessible charge states near the energy of the targeted defect will provide a channel for ionization and decoherence. When identifying good spinful defects for quantum coherence, a consideration of the undesirable defects in the material, through simulation and measurement, will help clarify if the defect is likely to be stable within that material. If the charge state is stable, and the defect is designed without orbital degeneracy, then the only effect of an applied electric field would be to change the wave-function composition of the ground-state eigenstates,[106] which leads to an electric-field-dependent spin precession frequency for the NV center.[104] The spin–orbit interaction drives the wave-function composition change, so in a light element solid such as diamond, the overall strength of the spin–orbit interaction is very weak; thus electrical noise from the surrounding material can be small. As a result, materials made from light elements are very attractive for potential defect hosts; these often include oxide materials that also have wide bandgaps. For electrical qubit devices based on silicon, the exchange and orbital energies cannot yet be made so high, so the operating temperature must be much lower; however, the larger spin–orbit interaction permits electrical control of the spin, for example, through electric dipole magnetic resonance (EDMR). Similar constraints are present for silicon qubits to diamond qubits, but at temperatures two orders of magnitude smaller. As the defect wave-function radii for silicon are much larger, the level of purity required for functional qubits is much higher in silicon. An alternate approach is to focus on deep levels in silicon, where the defect wave functions are correspondingly smaller. These deep levels, however, are often of lower symmetry (e.g., the T center[107]) than simpler substitutional defects and could involve complexes. A major materials challenge is learning how to preferentially form these complexes without creating a sea of other defects around them that provide magnetic and electric noise.

Another major source of noise acting on these centers is the spin noise; here the high formation energy of defects in diamond (especially intrinsic defects) suppressed the presence of the spin noise sources. Furthermore, the wide gap of diamond suppresses the exchange interaction between nearby spins,[108] limiting the interaction on scales less than a couple of nanometers to the much weaker dipolar interactions. The most significant sources of spin noise in diamond appear to be substitutional nitrogen, the so-called P1 centers. Similarly, near the surface, the presence of disorder appears to substantially reduce the spin coherence times of nearby spin centers.[109–117] This is a significant challenge for nanoscale sensing, which requires coherent spins located very near the surface of the material. Electric field effects also broaden the optical line, which can be mitigated by depleting carriers locally.[70,72] Finally, electron–nuclear interactions are very significant in materials with endemic spins; here the low natural abundance of nuclear spinful isotopes for carbon (for NV's) and for silicon (for silicon spin qubits) favors long spin coherence times for spin centers. For electrical devices based on silicon, the much smaller host bandgap allows qubit–qubit interactions based on exchange, as the exchange interaction extends over much larger distances than in diamond.[14,108]

Two speakers continued the discussion by describing how (1) dynamically controlling the spin and charge environment around an NV sensor can be used to improve their utility for sensing (P. Maurer), and (2) how similarly controlling the hyperfine (nuclear) environment around silicon spin qubits can stabilize them for quantum computation (A. Laucht).

Maurer focused on engineering the noise environment for qubit sensors with applications to sensing: Quantum sensing relies on highly coherent qubits to probe magnetic[101,102] and electric[104] fields, temperature,[118] and pressure[119] with nanoscale spatial resolution. Qubit sensors based on nitrogen-vacancy (NV) centers in diamond are point-like probes with optically detectable electron spins at ambient and cryogenic conditions. This has enabled applications ranging from mapping vortex structures in superconductors and magnetic properties of 2D materials[120] to probing magnetotactic bacteria[121] and cellular organization in embryogenesis.[122] These applications rely on the high sensitivity, enabled by long spin coherence times, and the ability to position the NV probe within the nanoscale vicinity of a desired target.

The sensitivity of these quantum sensors scales with the square root of the qubit coherence time.[123] For nanoscale sensing, the qubit sensor usually needs to be brought into close proximity with the sensing target, which means that dephasing due to surface noise becomes an important factor. Hence, understanding and engineering electric and magnetic noise sources associated with the diamond surface has become a central factor in extending the coherence of diamond qubit sensors and, with that, the range of potential applications.

Two orthogonal, but complementary, strategies have been successfully explored to drastically increase coherence for near-surface NV centers. The first approach relies on material engineering to create surfaces with a reduced density of charges traps and spins. This can be achieved by controlling the surface termination through carefully designed annealing steps and limit exposure to crystallographic damage during processing.[116,124] Alternatively, surface defect occupation and their dynamics can also be controlled by engineering dielectric constants of the surrounding materials[125] and/or through the creation of heterostructures that lead to a depletion of surface defects.[126] These approaches have shown to substantially increase qubit

coherence by as much as a factor of 100. The second approach relies on dynamical decoupling to reduce the adverse effects of surface noise on the spin qubit. Specifically, many sources of surface noise are highly non-Markovian, enabling an efficient use of dynamical decoupling. Using a Carr–Purcell–Meiboom–Gill sequence with up to 1000 microwave π pulses, coherence times for near-surface NV centers can be extended from a few microseconds to values close to 100 μs.[125] These dynamical decoupling techniques enable the reconstruction of the noise power spectrum experienced by the NV center.[127] Connecting these experimentally observed noise power spectra with established materials characterization techniques, such as x-ray photoelectron spectroscopy (XPS), near-edge x-ray absorption fine structure (NEXAFS), and ultraviolet photoelectron spectroscopy (UPS), could enable a deeper understanding of the microscopic mechanisms responsible for NV dephasing at interfaces.

The second speaker in this session, A. Laucht, reported on microwave-induced electron spin resonance frequency shifts in silicon spin qubit devices. Spin qubits in silicon have shown great promise as the platform to host a quantum processor, as evidenced by their high (>99%) 1- and 2-qubit gate fidelities.[1–3,128–130] In order to ensure the viability of this platform when scaling to greater numbers of qubits, it is important to fully understand and characterize the noise sources influencing these devices at the microscopic level.

One of the most prevalent noise sources in silicon spin qubit devices is magnetic noise, caused by the flip-flopping of nonzero spin isotopes of silicon, such as ^{29}Si, that are present in the vicinity of the qubit. This source of noise has been significantly reduced in recent years with the advancement of isotopic enrichment, which has resulted in the quantity of ^{29}Si present in qubit devices being reduced from approximately 47,000 ppm, down to only 50 ppm. At such low ^{29}Si concentrations, the qubit no longer experiences a net fluctuating magnetic field from the surrounding nuclei, but rather is strongly hyperfine-coupled to only a few individual ^{29}Si nuclei in the qubit's vicinity.[131,132] This has two benefits: first, the strong coupling of these ^{29}Si nuclei allow for their accurate characterization and, hence, the ability to initialize the nuclei into some desired state, or even use them as a qubit in their own right[133] or as quantum memory.[134] The second benefit is that the strong hyperfine coupling between the qubit and the individual ^{29}Si has the effect of detuning the ^{29}Si nuclei from one another in energy. With a sufficient degree of detuning, such that the detuning is greater than the coupling between the nuclei, the flip-flopping between the nuclei is suppressed, resulting in the elimination of magnetic fluctuations experienced by the qubit. This is an effect known as nuclear freezing.[135] However, if this hyperfine coupling is removed, for example, through the physical removal of the electron during readout, then the spin bath is "unfrozen" and fluctuations of the strongly coupled ^{29}Si nuclei can manifest as non-Markovian noise experienced by the qubit.

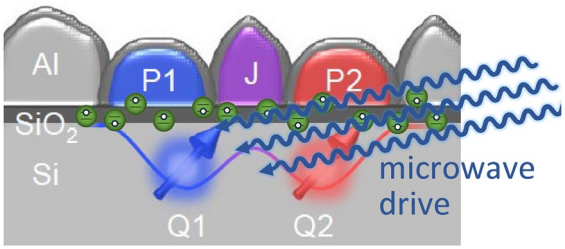

Figure 5. Excitation of two-level fluctuators via microwave drive. Two-level fluctuators in the gate oxide get excited via the same microwave drive that is used to control spin qubits in silicon.[136] This can lead to a shift in the wave function of the electron and consequently to a change in its *g*-factor and resonance frequency, resulting in contextual noise during the measurement. This effect has been observed in ^{31}P donor spin qubits, as well as in SiMOS and SiGe quantum dot spin qubits.

There is, however, another noise source observed in these devices, which is contextual in nature (see **Figure** 5); for example, it depends on how the measurement is performed: Noise originates from a shift in the resonance frequency of the spin, induced by the application of the oscillating pulse used to control the qubit, either at radiofrequency or microwave frequency. This pulse-induced resonance shift has been independently observed in a number of devices from groups around the world.[4,134,137–140] Although the exact characteristics of this effect differ depending on the exact nature of the device used, there appear to be unifying features observed across the different silicon platforms such as donors and quantum dots in Si/SiGe and SiMOS:

1. The resonance frequency shift occurs over some nonzero time scale after the application of a control pulse and saturates within a few microseconds. The resonance frequency returns to its original value over hundreds of microseconds, after the pulse has ceased. The finite time scale of this resonance shift differentiates this effect from AC Stark shift, which is instead an instantaneous shift in frequency during the application of an off-resonant pulse.
2. The magnitude of the resonance frequency shift depends on the power and duration of the pulse and does not require the pulse to be in resonance with the qubit.
3. The resonance shift appears to be affecting the Zeeman term in the qubit Hamiltonian, with some evidence to suggest that the effect is caused by a change in the gyromagnetic ratio of the spin.
4. This effect leads to a reduction in the coherence of the qubits and can even be the dominant source of infidelity in gate operations.[4]

The time scale over which this resonance shift occurs, along with the dependence on the power and duration of the applied pulse, potentially indicate that this is a temperature-related effect, caused by heating of the device with the application of a control pulse.[139] However, further investigation by the team

at UNSW revealed a nontrivial dependence of the magnitude of the pulse-induced frequency shift on device temperature and applied magnetic field, indicating that a more complex phenomenon than a direct temperature dependence could be at play.

Although a microscopic understanding of this effect is still lacking, there are two prevalent theories regarding its origin. The first theory is that the oscillating drive results in local heating of the device, which in turn, leads to a variation in strain that affects the conduction-band edge.[139] The second theory is that the drive instead leads to the excitation of charge traps and two-level fluctuators in the gate oxide, which in turn, leads to a variation in the electrostatic environment. Both of these effects ultimately result in the deformation of the electronic wave function and, hence, lead to a change in the gyromagnetic ratio through spin–orbit coupling.[141,142] Experimentally, it is difficult to tell them apart using standard experimental methodology. Systematic temperature-dependent studies as well as tests of different materials could be required.

Looking to the future, there are a number of potential solutions that can be implemented to mitigate this effect. In the short term, a number of independent research groups applied an off-resonant "saturation pulse" at the beginning of each measurement.[4,137,138] This pulse is applied at a sufficient power and duration such that the resonance frequency shift is saturated, resulting in no further frequency shift occurring through the application of subsequent control pulses. This, however, is not a scalable technique and has the disadvantage of lowering the coherence of the qubits. A medium-term solution is to attempt to mitigate the effect of strain and/or charge traps through careful choice of high-quality materials. A more long-term solution, however, could instead focus on a shift in paradigm from spin states as computational basis states with local, pulsed control fields to driven/dressed states as computational basis states with always-on global control fields.[143–148] Such implementations have the benefit of not only decoupling the qubits from magnetic noise along the quantization axis of the qubit, but the always-on control field also offers a method of saturating any pulse-induced resonance shifts, mitigating this contextual error source.

More recently, there has also been a detailed experimental study conducted in Reference 149 that shows that raising the operation temperature of the qubits can reduce their sensitivity to these microwave pulses. Furthermore, a theoretical study presented in Reference 136 shows that the main features of this phenomenon can be explained by two-level systems if considered to form an interacting random-field glass.

Predictive challenges for atomic-like quantum systems

The final round of discussions was led by L. Bassett, who started with an overview of criteria for point-defect searches that could yield qubits with better properties and new functionalities.

Much of our understanding of the physics and applications of defect-based qubits is based on work on the diamond NV center. Certainly, the diamond NV center has special properties, including its robust spin coherence along with room-temperature spin initialization and readout. However, we now know that the diamond NV center is not unique, and that other defects may be better suited for certain applications. In recent years, it has been demonstrated that the diamond silicon-vacancy (SiV) can be employed to efficiently generate coherent optical photons even in nanophotonic devices, addressing a problem of optical coherence for NV centers close to surfaces that had previously blocked progress in realizing efficient light–matter interfaces.[150,151] Improved photon coherence is attributed to the decrease in sensitivity to electric fields due to inversion symmetry of SiV centers. Much of that work has focused on the negatively charged SiV center; however, other Group-IV-vacancy defects such as GeV and SnV are poised to play a role, as are other charge states.[152]

Furthermore, SiC is a promising system for atom-scale spintronics as it exists in numerous polytypes and hosts a variety of spin defects. Koehl and co-workers showed that defect spin states in the 4 H polytype of SiC (4 H-SiC) can be optically excited and coherently controlled.[153] Additionally, it was demonstrated that 4 H-SiC could accommodate single point defects, which can be coherently controlled with long spin coherence times at room temperature.[154] Divacancies in 3C- and 4 H-SiC are NV center analogous, and a robust spin-to-photon interface was revealed in both SiC polytypes, which could enable a high-fidelity preparation and readout employing resonant infrared light.[155] Diamond NV centers have been studied extensively in the last decades for application in the field of quantum information science, and are foreseen to have a continued interest. They still serve as a benchmark for developing analogous systems, such as the single- and divacancies in SiC.

Building on these examples, there is enormous potential to identify point defects in new materials that are optimized for particular applications. For example, it could be important to select a particular emission wavelength, for example, in the telecommunications band for long-distance transmission through fiber, or in the near-infrared biological window for biological sensing applications. Similarly, for spin–photon interfaces, it is important to maintain both spin and optical coherence, whereas for room-temperature quantum sensors, the primary metric could be the signal-to-noise ratio for incoherent optical spin readout. For designing novel point defects, a combined approach from experiment and theory is required; a proposed approach is by starting with the desired application goals, to ascertain the requirements in order to classify a range of crystal hosts and defects with the prescribed properties, and only then initiate targeted research to engineer the materials and improve experimental techniques.[60] From a theoretical point of view, the ability to quantitatively predict and calculate such complex, application-specific properties, including room-temperature,

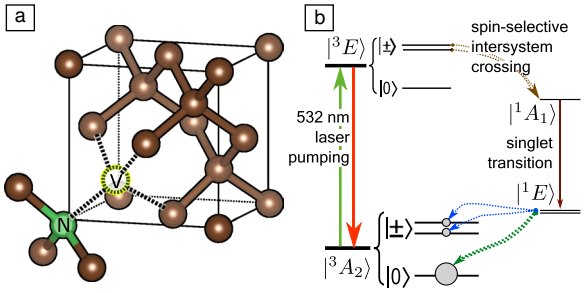

Figure 6. Schematic diagram of the optical spin polarization of a nitrogen-vacancy center in diamond. (a) Structure of nitrogen-vacancy defect in diamond. (b) The schematic level diagram is typical at elevated temperatures for the ^3E excited state. The $m_S = \pm 1$ states could split by either strain, electric fields, or constant external magnetic fields. Optical spin polarization starts with spin-selective intersystem crossing (brown curved arrows) from the ^3E triplet excited state toward the $^1A_1$ singlet excited state upon illumination. The electron decays to the ^1E singlet shelving state where the dominant relaxation path is toward the $m_S = 0$ state of the $^3A_2$, so that level will be predominantly populated after several optical cycles as depicted by the large circle. The dominant spin-selective route (green arrow) between ^1E and $^3A_2$ is caused by strong electron–phonon coupling between $^1A_1$ and ^1E.

spin-dependent optical contrast, has only recently become possible. The two speakers in this session described the current state of the art and outstanding challenges for this goal.

The first speaker in this session, A. Gali, focused on the physics of defect qubits, in particular, on the role of intersystem crossing (ISC) as a critical phenomenon and a topic of continued interest in research. The full optical spin-polarization process in an NV center comprises an ISC mechanism, characterized by nonradiative transitions between states of different spin multiplicity (i.e., between the singlet states and triplet states [see **Figure** 6]).

The ISC mechanism between the singlet states and the ground-state triplet state in diamond NV centers can be understood by strong electron–phonon coupling between the singlet states, which is a combination of pseudo Jahn–Teller and damped dynamic Jahn–Teller effect.[156] Moreover, the temperature dependence of the ISC rate toward the ground-state triplet (i.e., the lifetime of the shelving singlet state) can be understood using a Herzberg–Teller type of ISC mechanism, where the vibronic picture enables to link the shelving singlet state to the triplet ground state.[156] The vibronic or polaronic states also play an important role in the ISC process between the excited state triplet and the singlet states.[157] The strong electron–phonon coupling is reflected in the temperature dependence of the fluorescence spectrum too, where the fine structure disappears at T >20 K.[157,158]

These results highlight the importance of electron–phonon coupling for the properties of defect qubits that frequently occurs due to degenerate electronic states. As a consequence, *ab initio* modeling of defect qubits often requires to go beyond the Born–Oppenheimer approximation.[159] A very special case is the G-center in silicon,[160] a single-photon source[161,162] with optically read electron spin,[163] where the fine structure of the excited state in the GHz energy region can be understood as the fast rotational motion of the silicon interstitial in the defect.[164] This defect has low symmetry but the electron–phonon coupling is still an essential feature in understanding its fluorescence.

In particular, electron–phonon coupling is responsible for the temperature dependence of the longitudinal spin-relaxation time (T_1 time). Until recently, this issue could be discussed at phenomenological level (e.g., Reference 59). Due to the advance of the accurate description of defect spins from *ab initio* simulations, the temperature-dependent T_1 time has been recently studied for the diamond NV center at a microscopic level.

The spin-relaxation rate ($\Gamma = 1/T_1$) could be expressed as

$$\Gamma = \Gamma_0 + \Gamma_1^{(1)}(T) + \Gamma_1^{(2)}(T) + \Gamma_2^{(1)}(T) + \ldots, \qquad 1$$

where the superscript refers to the order of the spin-phonon interaction (i.e., terms with superscript 1 or 2 are linear or quadratic in the atom displacements, respectively) and the subscript refers to the order in perturbation theory. Γ_0 is a sample-dependent constant term arising from spin–spin interactions that is not discussed further. $\Gamma_1^{(1)}$ describes the absorption or emission of a single resonant phonon by the spin. Because the zero-field splitting energy of the NV ground-state triplet is small in comparison to typical phonon energies in diamond, this process is only relevant at sub-Kelvin temperatures.[165] $\Gamma_2^{(1)}$ corresponds to the Raman scattering of low-energy acoustic phonons via first-order interactions. However, it has been recently shown[166] that the first-order spin–phonon interactions provide only negligible contributions to Raman scattering for the NV center in diamond. The major effect comes from $\Gamma_1^{(2)}(T)$ (i.e., the quadratic displacements of ions). This has been confirmed by another study,[167] which also applies to the negatively charged boron vacancy in hBN. *Ab initio* theory should be applied to understand the strongly anisotropic spin-relaxation time for the neutral silicon-vacancy (SiV^0) centers in diamond,[152] where it is still not clear whether the metastable singlet state or quasilocal vibration modes play a crucial role in the process.

SiV^0 is also mentioned from another important perspective. By combining experiments and *ab initio* calculations, it was proven that optically detected magnetic resonance (ODMR) of the spin triplet ground state could occur via bound exciton states.[168] Bound exciton states associated with the effective mass state with hydrogenic excitation series may be not so commonly known in the quantum optics community coming from atomic physics. These excited states often occur in small bandgap semiconductors, but can be present also in wide bandgap materials such as diamond or 4 H-SiC. From an *ab initio* simulation point of view, the description of the bound exciton states is a great challenge because of the spatial extension of the respective wave function building up the bound exciton states[169] that may be treated using a size scaling method with extrapolation to the dilute limit.[168]

It is noted that there is great progress on *ab initio* simulations of the dynamics of the defect spins in realistic spin bath environments,[169] which can provide spin dephasing and spin coherence times. The spin dynamics of the diamond NV center,[170–172] SiC defects,[73,170] and boron vacancy in hBN[64,173–176] have been thoroughly described via coupled cluster expansion methods.[177–179] As a next step, the spin–spin interaction associated with realizing qubit gate operations may be studied in the presence of nuclear or electron spin baths to find optimal quantum optics protocols and to guide materials engineering.

The second speaker in this session, Y. Ping, presented first-principles calculations of the photophysics of quantum defects, in particular in 2D materials. Point defects in 2D materials are prominent candidates for quantum technologies.[180] There are several practical advantages, for example, allowing easier integration into smaller solid-state devices because of their atomically thin thickness, possibility for applying large strain, easier manipulation of defects through scanning techniques, and tunability through varying substrates. In particular, point defects in hBN have shown promising room-temperature single-photon emission[181] and optical contrast in ODMR,[182,183] which points to its potential in single-photon emitters (SPEs) and spin qubits. Defects in transition-metal dichalcogenides (TMDs) are also considered promising candidates, given their much lower nuclei spin concentration than hBN (potentially longer nuclei spin coherence time), although their bandgaps are much smaller, which could lead to coupling between defect and bulk states of the host materials.

Although numerous candidates in 2D materials have been suggested as single-photon emitters (SPEs) or spin qubits, the most known experimentally confirmed spin defect in hBN is the negatively charged boron vacancy (V_B^-),[175,184] which does not show superior optical properties for quantum applications. For example, the zero-phonon line (ZPL) in the photoluminescence (PL) spectrum is not prominent, accompanied by broad phonon sidebands, in sharp contrast to the dominant ZPL peaks of SPEs in hBN. This indicates that the photoluminescence (PL) lifetime is largely determined by nonradiative processes, in another word, it is not a good SPE intrinsically. The other potentially promising candidates such as carbon-based defects[183,185] have not been resolved with the exact chemical composition. Therefore, there is still critical demand for identifying promising SPEs and spin qubit candidates in 2D materials. Important criteria for designing new spin qubits include a stable high spin state, as unpaired electrons are necessary to use the spin as a qubit, sizable zero-field splitting, which is critical to isolate the $m_s \pm 1$ and 0 sublevels even at $B = 0$ T. Furthermore, bright optical transitions allow for optical preparation and readout of spin qubits, and possible single-photon emission. Finally, accessible intersystem crossing (ISC) is critical for pure spin state initialization and readout as shown in **Figure** 7a.

Anisotropic and weak dielectric screening in 2D materials leads to large many-body effects such as electron–hole

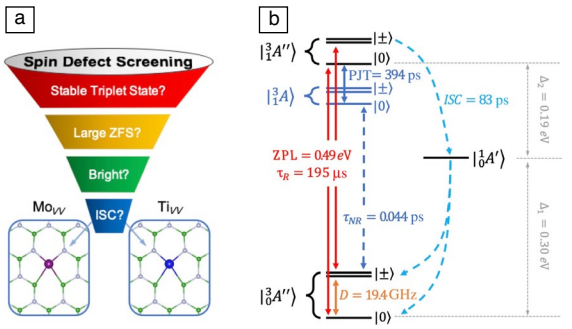

Figure 7. Screening of spin defects in semiconducting solids. (a) Schematic of screening criteria and a workflow, where we first search for defects with stable triplet ground state, followed by a sizable zero-field splitting (ZFS), then "bright" optical transitions between defect states required for single-photon emitters (SPEs) or qubit operation by photon, and at the end large intersystem crossing (ISC) rate critical for pure spin state initialization and readout. At the bottom, two candidates of Ti_{VV} and Mo_{VV} in hBN were found to be promising based on such criteria. (b) Multiplet structure and related radiative and nonradiative recombination rates of the Ti_{VV} defect in hBN as the candidate obtained from computational screening, computed at $T = 10$ K. The radiative process is shown in red with zero-phonon line (ZPL) and radiative lifetime (τ_R); the ground-state nonradiative recombination (τ_{NR}) is shown as a dashed line in dark blue; and finally, ISC to the singlet state from the triplet excited state is shown in light blue. The zero-field splitting (D) is denoted by the orange line. For the Ti_{VV} defect, the pseudo-Jahn–Teller (PJT) process is shown with a solid line in dark blue.[193]

interactions that impose complexity and outstanding challenges in *ab initio* 2D defect calculations.[186,187] Ping discussed recent developments in her group of calculating important physical parameters for excited states of charged defects in 2D materials, using first-principles many-body perturbation theory.[186] In particular, the charged defect energy levels are calculated using the *GW* approximation for accurate electron correlation with a proper 2D charged defect correction that avoids spurious charge interactions.[188,189] The radiative lifetime, computed by solving the Bethe–Salpeter equation, includes excitonic effects, which is critical for 2D materials.[190] Nonradiative lifetimes include explicit electron–phonon coupling at 2D defects through the effective phonon approximation,[191] which has shown success for nonradiative rates of 3D semiconductors.[192] Combining radiative and nonradiative lifetime provides the photoluminescence (PL) lifetime, which can be compared to experimental PL measurements directly. Finally, the ISC rate includes electron–phonon coupling and spin–orbit coupling computed from time-dependent DFT.[193] With all the information of energy levels and kinetic rates computed above, the multistates diagram can be constructed and spin-dependent PL contrast (ODMR contrast) can be computed. Specifically, by solving a kinetic master equation with computed rates as inputs, under pump light and external magnetic/microwave fields, the excited state occupation of different spin states can be determined, which gives ODMR contrast as a key parameter for optical readout fidelity. Significant challenges remain for intersystem crossing rate calculations,

where strong electron correlation of excited state, (dynamical) Jahn–Teller distortion, and spin–orbit coupling need to be calculated accurately simultaneously, which is extremely difficult computationally. A combination of group theory analysis, DFT, and high-level quantum chemistry method may be necessary to solve the problem satisfactorily.

Ping then discussed an example of defect identification: the carbon dimer defect in hBN,[194] where PL lineshape, PL lifetime considering both radiative and nonradiative processes, Huang–Rhys factor, and ZPLs from simulations compare well to available experimental data simultaneously. Therefore, this defect is suggested to be a viable 2 eV SPE in hBN observed experimentally. Besides identifying experimental defects from *ab initio* calculations, Ping also illustrated an example of designing new quantum defects from *ab initio* calculations.[193] For example, the Ti substitution of divacancy, Ti_{VV}, in hBN shows fast ISC rate due to large spin–orbit coupling and electron–phonon coupling. Mo_{VV} shows reasonably fast ISC and shorter radiative lifetime than Ti_{VV}, which could be promising as optically addressable spin qubit (as shown in Figure 7b). Other examples are optically addressable point defects such as Si_{VV}, which show promising light-emission properties for single-photon emission.

Spin decoherence, however, is a major challenge in quantum information materials. At very low temperature and large B field, decoherence mainly arises from fluctuating magnetic fields due to nuclear spin flip-flop transitions. At finite temperatures, other effects such as phonons, impurities, and electron–electron interactions through spin–orbit coupling can become dominant. Spin–orbit mediated spin-phonon relaxation in spin qubits can also result in short coherence time. Over the last few years, the Ping group has developed methodology based on *ab initio* density-matrix dynamics,[195,196] with quantum electron–phonon, electron–electron, and electron-impurity scattering with self-consistent spin–orbit coupling. Ping showed the predicted spin relaxation (T_1) and ensemble dephasing times (T_2*) of conducting electrons or holes in disparate solids in excellent agreement with experiments, regardless of dimensionality and symmetry.[195–198]

Regarding open questions for spin relaxation T_1 and coherence time T_2 calculations of spin defects in solids, one of the remaining challenges is the numerical convergence of the electron–phonon calculation for defect supercells, in particular for couplings with low energy or delocalized phonons in spin defect T_1 calculations. The required supercell sizes for converging electron–phonon coupling and energy conservation at meV scale (spin splitting) can be enormous. Ping suggests that novel methods such as the embedding method of electron–phonon matrix elements could be a viable pathway to explore. Also, a fully first-principles approach to study T_1 for spin qubits under macroscopic confinement potentials in large-size nanostructures (e.g., tens of nanometer-size quantum dots) is unavailable. This is significantly beyond the computational feasibility of *ab initio* calculations. Ping suggests that combining model Hamiltonians for nanostructures with *ab initio* fitted parameters could be a promising pathway for spin T_1 in quantum dots, which realizes an ideal balance between computational cost and accuracy.

High nuclei spin concentration in hBN could be a major source for its spin decoherence at low temperature, possibly posing a fundamental limitation for hBN in quantum applications. Recent work has proposed to control and utilize nuclei spin in hBN through optically detected nuclear magnetic resonance (ODNMR) assisted by point defects, which opens a new frontier with nuclear spins in van der Waals materials for quantum information science.[173] Furthermore, recent progress has shown significantly elongated spin coherence time for V_B^- in hBN through dynamical decoupling techniques. They suppress magnetic noise and extend the spin coherence time by nearly two orders of magnitude, approaching the fundamental T_1 relaxation limit.[199]

Summary

M. Friesen pointed to the need for improved atomic-level growth control and simulation to allow for reliable qubit scaleup. C. Déprez highlighted the effect of hysteretic changes in the potential landscape on qubit stability and the need to further explore the origin of these hysterectic effects and to develop control mechanisms. C. Anderson highlighted the role of acoustic, magnetic, and electric noise sources and the particular influence of the proximity of interfaces on the stability of defect-based qubits. He also emphasized the need to have detailed knowledge of the structure and the Hamiltonian of the defect to be able to predict the influence of the various noise sources on the qubit operation. T. Zhong highlighted the fact that rare earth ions in solids possess properties, including long coherence times and narrow emission widths that make them leading candidates for quantum memories. However, he also explained that there are only a few demonstrations thus far of individual ions being operated as spin qubits and that considerable development is required in this area. P. Maurer highlighted the information that can be obtained about the influence of the environment on a qubit through operation of the qubit as a sensor and that this can be used to work out better ways to isolate the qubit from its environment. A. Laucht highlighted the role of microwave-field-induced electron spin resonance frequency shifts on the performance of Si qubit devices and the need to develop suitable control mechanisms or to operate the qubits in a different mode, for example, shifting from spin states as the computational basis states to driven/dressed states with always-on control fields. A. Gali and Y. Ping both highlighted the enormous progress that has been made in the theoretical modeling of defect-based solid-state qubit candidates in recent years including the dynamics of the systems in realistic environments and that next steps involve modeling of spin–spin interactions during qubit gate operations and the need for greater focus on modeling of sources of decoherence.

Notably, specific materials suggestions will be application specific. When using qubits for sensing at room temperature, the phonon sideband can be used for readout and coherent emission at a fixed wave length is not important. The readout contrast, spin-relaxation time, and coherence time are the most important parameters. Room-temperature operation likely restricts use of host materials with constituting elements in the second row of the periodic table (such as diamond, silicon carbide, and boron nitride) because the spin-relaxation time exponentially decays with the given effective phonon frequency occupied at the given temperature. High enough phonon frequency requires light elements in the crystal. For quantum communication, the coherent emission in ZPL should be strong, stable, and identical, independent from the environment. Quantum computation requires additional conditions such as well-controlled gate operations between qubits (that could pose stringent conditions for the positioning of the qubits in the solid) and high scalability.

A concerted materials science focus and push is needed in order for quantum computing, sensing, and communications to advance to the point that it offers true advantage. This would include both engineering means of identifying and tuning to sweet spots and designing Hamiltonians and fundamental materials innovations in growth and characterization to exploit these sweet spots. Only by combining both of these strategies which tune, understand, grow, and optimize quantum materials will ideal performance be unlocked through exquisitely engineered materials and interfaces that provide access to a broad range of available operating parameters. This is increasingly recognized by funding agencies, as evidenced by materials science focused efforts, for example, by the AFOSR. The contributions to this workshop identify some of the key areas of focus needed to make real progress for the particular qubits systems that were examined here. The Quantum Staging Group will continue to promote activities across a range of qubit platforms that bring practitioners in the field together to focus on the questions that need to be answered. The authors of this article aspire to the goal of providing insights to the materials community to solve underlying problems and find guidance in this work toward new collaborations and collaborators.

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Author contributions

A.S., C.C., B.S., and J.M.: provided an initial outline and plan for this manuscript. V.B.H. and A.S.: then used notes taken during the workshop itself to draft an initial version of this work. Each author then wrote the description of those parts of the workshop associated with their name as delineated in the text. All authors edited the entire manuscript for clarity and correctness.

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Data availability

No data sets were generated or analyzed during the current study. The code used to generate Figure 2 can be found at https://zenodo.org/records/10075090.

Conflict of interest

On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Adam Gali is a chief scientist at the Wigner Research Centre for Physics, the leader of the "Lendület" Semiconductor Research Group, the founder of Wigner Advanced Materials Integrated Laboratory and Optically Detected Magnetic Resonance Laboratory, a leader at the National Quantum Information Laboratory of Hungary, a professor at Budapest University of Technology and Economics, and Doctor of Science of the Hungarian Academy of Sciences. He studies the magneto-optical properties of point defects in semiconductors and semiconductor nanostructures by means of *ab initio* methods and experimental techniques, to develop efficient biomarkers, solar cells, and solid-state defect quantum bits. He recently has developed and implemented methods to fully characterize nitrogen-vacancy center in diamond from first-principles methods that can be applied to other solid-state defect quantum bits such as other vacancy-impurity defects in diamond, divacancy in silicon carbide, and other defects in three- and two-dimensional solids. Gali can be reached by email at gali.adam@wigner.hu.

André Schleife is a Blue Waters Associate Professor in Materials Science and Engineering at the University of Illinois at Urbana-Champaign. He obtained his diploma and PhD degree from Friedrich-Schiller-Universität Jena, Germany. He was a postdoctoral researcher at Lawrence Livermore National Laboratory before starting at the University of Illinois in 2013. He actively organizes national and international schools, workshops, and tutorials to advance the community around cutting-edge first-principles simulations of materials. His research focuses on efficient, fast, and scalable quantum mechanical simulations and their applications. He develops and uses predictive computational materials science techniques at the nexus of solid-state physics and modern data-driven approaches to study electronic materials, metals, and defects in materials with quantum information applications. Schleife can be reached by email at schleife@illinois.edu.

Andreas J. Heinrich is a researcher in the field of quantum-coherent nanoscience with a focus on quantum spins on surfaces. He pioneered spin excitation and single-atom spin resonance spectroscopy with scanning tunneling microscopes—methods that have provided high-resolution access to the quantum states of atoms and nanostructures on surfaces. He is fascinated by the world of atoms and nanostructures, built with atomic-scale precision, and educates the public on nanoscience as demonstrated by the 2013 release of the movie *A Boy and His Atom*. He received his PhD degree in 1998 from the University of Goettingen, Germany, where he also completed postdoctoral research. Heinrich spent 18 years in IBM Research. He became a distinguished professor at Ewha Womans University in Seoul, South Korea, in August 2016, and started the Center for Quantum Nanoscience (QNS) of the Institute for Basic Science (IBS) in January 2017. Heinrich can be reached by email at heinrich.andreas@qns.science.

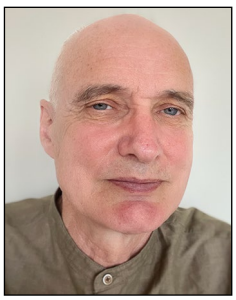
Arne Laucht is a Scientia Associate Professor for Quantum Engineering within the School of Electrical Engineering and Telecommunication at the University of New South Wales, Sydney, Australia, and the head of Quantum Measurement at Diraq Pty Ltd. He received his PhD degree in physics from Technische Universität München, Germany, in 2011. His main research interests include quantum physics, spin quantum computing, SiMOS quantum dots, dressed states, color centers, and nanoscale device engineering. Laucht can be reached by email at a.laucht@unsw.edu.au.

Bruno Schuler is the group leader of the 2D Quantum Materials Group in the nanotech@surfaces Laboratory at Empa—Swiss Federal Laboratories for Materials Science and Technology, interested in exploring low-dimensional quantum materials with cross-correlation scanning probe microscopy at the ultimate spatial, energy, and time resolution. He received his BSc and MSc degrees in physics from ETH Zürich, and his PhD degree and postdoctoral research were conducted at the IBM Research Laboratories in Zurich, where he pioneered chemical structure identification of single molecules using CO-tip noncontact atomic force microscopy. From 2016 to 2020, he worked as a Swiss National Science Foundation Postdoctoral Fellow and junior group leader at the Lawrence Berkeley National Laboratory's Molecular Foundry. In 2020, he received a European Research Council Starting Grant for developing an ultrafast terahertz-scanning tunneling microscopy for studying the dynamics in two-dimensional materials. Schuler can be reached by email at Bruno.Schuler@empa.ch.

Chitraleema Chakraborty is an assistant professor in materials science and engineering and physics at the University of Delaware. She received her PhD degree in materials science from the University of Rochester in 2018. In 2018, she joined the Research Laboratory of Electronics at the Massachusetts Institute of Technology as a postdoctoral research associate in quantum photonics. Following that, she joined the Computational Materials Science Group at Harvard University as a postdoctoral fellow. Chakraborty can be reached by email at cchakrab@udel.edu.

Christopher P. Anderson is currently an assistant professor of materials science and engineering at the University of Illinois at Urbana-Champaign. Previously, he was an intelligence community postdoctoral research fellow at Stanford University. He received his PhD degree in physics from The University of Chicago, and graduated with a BS degree in both physics and chemistry from the University of Michigan. His research focuses on developing the materials and devices that will enable the next generation of quantum information technologies. Specifically, he explores how photonics and solid-state systems can be used in long-distance entanglement distribution, quantum sensing, and in modular quantum computing. Anderson can be reached by email at cpand@illinois.edu.

Corentin Déprez is a postdoctoral researcher at QuTech/Delft University of Technology, The Netherlands. He completed his PhD degree at Institut Néel, France, where he researched quantum Hall physics and electron interferometry in graphene heterostructures. His current research focuses on hole spin qubits in planar germanium heterostructures and developing new strategies for the operation of spin-qubit arrays. Déprez can be reached by email at C.C.Deprez@tudelft.nl.

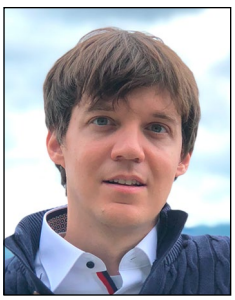


Jeffrey McCallum is a professor in the School of Physics at the University of Melbourne, Melbourne, Australia. He received his PhD degree from The University of Melbourne in 1989. His current research interests focus on ion implantation approaches for quantum device formation in solid-state systems with applications in quantum computing, communications, and sensing. He also focusses on defect characterization and control. McCallum can be reached by email at jeffreym@unimelb.edu.au.

Lee C. Bassett is an associate professor of electrical and systems engineering at the University of Pennsylvania. Following undergraduate studies at The Pennsylvania State University (BS physics, 2004), he traveled to the United Kingdom as a Marshall Scholar and National Science Foundation Graduate Research Fellow to complete the MASt (Part III) in mathematics (2005) and PhD degree in physics (2009) at the University of Cambridge, UK. From 2009 to 2013, he was a Hewlett Packard Postdoctoral Fellow at the University of California, Santa Barbara. His research interests include quantum optics, spins in semiconductors, nanophotonics, and control of complex quantum systems. Bassett can be reached by email at lbassett@seas.upenn.edu.

Mark Friesen is a distinguished scientist of physics at the University of Wisconsin–Madison. He received his PhD degree in physics in 1995 from the University of Minnesota. His current research focuses on the development, application, and materials science of solid-state qubits, especially in quantum dots. Friesen can be reached by email at friesen@physics.wisc.edu.

Michael E. Flatté is a professor in the Department of Physics and Astronomy at The University of Iowa (UI). He received his AB degree in physics from Harvard University in 1988, and his PhD degree in physics from the University of California, Santa Barbara, in 1992. After his postdoctoral work at the Institute for Theoretical Physics, University of California, Santa Barbara, and the Division of Applied Sciences, Harvard University, he joined the faculty at UI in 1995. He has more than 270 publications and 10 patents. He has an adjunct appointment as a professor in the Department of Applied Physics, at Technische Universiteit, Eindhoven, The Netherlands. His research interests include optical and electrical control of spin dynamics in materials, novel spintronic devices, quantum sensors, and solid-state realizations of quantum computation. He is a fellow of the AAAS and the American Physical Society. Flatté can be reached by email at michael-flatte@uiowa.edu.



Peter Maurer is an assistant professor at The University of Chicago. He completed his postdoctoral training at Stanford University, and received his PhD degree from Harvard University. His group combines quantum engineering, biophysics, and material engineering to develop biocompatible quantum sensing tools and apply them to real-world problems in the life sciences. Maurer can be reached by email at pmaurer@uchicago.edu.

Susan N. Coppersmith is currently a Scientia Professor and the head of the School of Physics at the University of New South Wales, Sydney, Australia. She has been working to develop quantum computers using quantum dots in silicon/silicon-germanium heterostructures since 2001. She is a Fellow of the Australian Academy of Science, the AAAS, the American Academy of Arts & Sciences, and the National Academy of Sciences. Coppersmith can be reached by email at s.coppersmith@unsw.edu.au.

Tian Zhong is an assistant professor in the Pritzker School of Molecular Engineering at The University of Chicago. He received his BEng degree from Nanyang Technological University, Singapore, in 2007, and his PhD degree in electrical engineering (physical engineering) from the Massachusetts Institute of Technology in 2013. From 2014 to 2017, he worked as a postdoctoral scholar at the California Institute of Technology on the topic of quantum memory and spin-photon interface based on rare earth doped crystals. His current research interests include quantum photonics and quantum interconnect technologies with a focus on solid-state quantum light–matter interfaces. He has more than 15 years of research experience in quantum science spanning quantum optics, quantum communication, coherent quantum materials, and nanophotonic quantum devices. Zhong can be reached by email at tzh@uchicago.edu.

Vijaya Begum-Hudde is currently a postdoctoral researcher in the Department of Materials Science and Engineering at the University of Illinois at Urbana-Champaign. She obtained her MSc degree in materials science and simulations from the Ruhr-Universität Bochum, Germany, and PhD degree in physics from the Universität Duisburg-Essen, Germany. Her doctoral thesis focused on the theoretical study of the spectroscopic properties of oxides with many-body perturbation theory. Currently, she is working on quantum embedding to study near-surface vacancies in oxides and is engaged in solving these problems with the aid of quantum computers. Her research interests include theoretical modeling of light–matter interactions, development of quantum algorithms and error mitigation, and exploring prospective materials for qubits. Begum-Hudde can be reached by email at vijayab@illinois.edu.

Yuan Ping is an associate professor at the University of Wisconsin. She received her BSc degree from the University of Science and Technology of China in 2007, PhD degree from the University of California, Davis, in 2013, and materials postdoctoral fellow from the California Institute of Technology in 2016. She was tenured at the University of California, Santa Cruz (2016–2023). Her research group focuses on developing and employing first-principles many-body theory and quantum dynamics for materials applications. She is a recipient of the Alfred P. Sloan Research Fellowship, National Science Foundaion CAREER Award, Air Force Yuoung Investigator Program Award, and American Chemical Society COMP OpenEye Award. Ping can be reached by email at yuanping@ucsc.edu.