Atomic-precision advanced manufacturing for Si quantum computing

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A materials synthesis method that we call atomic-precision advanced manufacturing (APAM), which is the only known route to tailor silicon nanoelectronics with full 3D atomic precision, is making an impact as a powerful prototyping tool for quantum computing. Quantum computing schemes using atomic (^{31}P) spin qubits are compelling for future scale-up owing to long dephasing times, one- and two-qubit gates nearing high-fidelity thresholds for fault-tolerant quantum error correction, and emerging routes to manufacturing via proven Si foundry techniques. Multiqubit devices are challenging to fabricate by conventional means owing to tight interqubit pitches forced by short-range spin interactions, and APAM offers the required ($\text{\AA}$ -scale) precision to systematically investigate solutions. However, applying APAM to fabricate circuitry with increasing numbers of qubits will require significant technique development. Here, we provide a tutorial on APAM techniques and materials and highlight its impacts in quantum computing research. Finally, we describe challenges on the path to multiqubit architectures and opportunities for APAM technique development.

Introduction

Leading quantum computation schemes are scaling up to system sizes and fidelities sufficient to outperform classical computers at certain tasks (e.g., quantum simulation), with some schemes achieving quantum advantages in certain cases.^{1–3} Even so, their future is rich with significant technological hurdles related to cooling, integrated control, and realization of encoded logical qubits using quantum error correction.⁴ To address scale-up challenges, Si schemes using spin qubits hosted either by dopant atoms (Kane)⁵ or quantum dots (Loss and DiVincenzo)⁶ are appealing owing to very promising qubit properties and the potential to integrate into dense nanoelectronics leveraging proven scalable Si foundry manufacturing.^{7–10} Quantum dot qubits are described in the separate papers by Gyure et al.¹¹ and Scappucci et al.¹² in this issue. For additional background on both dopant and quantum dot schemes, see the review by Zwanenburg et al.³

Figure 1a shows Kane-like dopant qubits in metal oxide semiconductor (MOS) stacks, common to Si nanoelectronics.

Typically, Si electronics are doped using Group III and V elements binding unpaired electron or hole spins, S , to harness as qubits. The binding energy is shallow (40–160 meV), allowing charges to move with voltages compatible with nanoelectronics to mediate qubit dynamics. Unpaired electron or hole spins can be used to access the nuclear spin, I , whose properties vary by dopant species and isotope, but generally host qubits with some of the longest quantum coherence times in the solid state.¹³

A conceptually straightforward spin qubit scheme uses ^{31}P donors, where $S=I=1/2$ (Figure 1b). Two important metrics for qubit information retention are the relaxation time, T_1 , which is a timescale for a qubit to exchange energy with its environment, and the dephasing time, T_2^* , which is a timescale for a qubit superposition state to evolve (decohere) to an unintended state due to quasi-static variation in the qubit's environment.³ Pointing to robust quantum information retention for computing, P has outstanding nuclear and e-spin relaxation ($T_{1n} > 100$ s, $T_{1e} > 30$ s) and dephasing times ($T_{2n}^* \sim 0.57$ ms, $T_{2e}^* \sim 0.27$ ms) within Si nanoelectronic devices.¹³ Long T_1 is

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due to negligible spin–orbit coupling and absence of piezoelectric phonons in Si. Optimal T_2^* is achieved by qubits in isotopically enriched ^{28}Si ($I=0$), to suppress background magnetic noise from 4.7% ^{29}Si ($I=1/2$) in natural Si. T_2^* is proving ample relative to qubit gating and readout (typically microseconds or less) for long gate queues required for computation.

The first dopant qubits were achieved by foundry methods, when Jamieson and Morello et al. demonstrated single-dopant qubits by ion implantation into MOS nanoelectronics, similar to Figure 1a.⁷ Morello’s qubits consist of a dopant atom tunnel-coupled to a nearby charge reservoir, for example

currently a hurdle to scale up dopant circuitry. Spin–spin dipolar coupling could serve to mediate slow (ms) two-qubit gates; however, the stronger exchange interaction provides more desirable speed (GHz) for gates.¹⁰ Exchange is proportional to electronic wavefunction overlaps, as illustrated in Figure 2b–d, and hence hypersensitive to donor separation, due to both the tiny dopant wavefunction ($r \sim 2.5$ nm) and valley-orbit-induced interference effects.^{14,15} The Voisin et al. article in this issue¹⁶ introduces the challenges. For implant, multiqubit circuitry is complicated by inherent positional disorder (straggle). Realistic straggles ~ 3 – 10 nm correspond to order-of-magnitude

variation of exchange coupling. Implant progress is focusing on mitigating straggle with thin oxides, low-energy implant, and integration with scanning probe ion apertures.¹⁷

An atomic-precision doping method that mitigates positional disorder is opening pathways to systematic studies of multiqubit interactions via control at the relevant interaction scales (Å). Originally proposed by Lyding and Shen, the technique positions single atoms in Si crystals with atomic precision via scanning tunneling microscope (STM)-assisted surface chemistry.^{18,19} This is accomplished by opening reactive sites on a hydrogen-terminated Si surface with STM, at which dopant precursor (e.g., PH_3) molecules react selectively, and decompose into isolated dopants (the so-called “hydrogen depassivation lithography (HDL)”). Researchers led by M. Simmons have demonstrated atomic-precision dopant spin qubits and recently two-qubit gates.^{20–22} A contemporary

version of the method, which we refer to as APAM, is described in **Figure 3**.²³

Here, we review impactful results, salient challenges, and emerging opportunities for APAM for quantum devices. We start by describing APAM methods and materials, then leading technology demonstrations, currently at the one- and two-qubit scale. Learning from these systems clarifies challenges to building larger circuits. We review one state-of-the-art architecture proposal, and its materials and fabrication challenges.²⁴ Finally, we describe some key challenges and opportunities to broaden APAM techniques.

Atomic-precision methods, materials, and structures

A central function of spin qubit nanoelectronics is to translate spin into measurable electrical signals and vice versa. In APAM devices, this is achieved by 2D precision-doped nanoelectronics (**Figure 1c**), comprising features with shapes and sizes controlled by atomic-precision templates.²⁵ Features range from mesoscale delta-doped sheets down to single atoms with ± 1 lattice-site precision, at ultra-high dopant densities $>10^{14} \text{ cm}^{-2}$ not achieved by other methods.²⁰ APAM structures are encapsulated in epitaxial Si, isolating them from interface disorder. APAM doping remains Ohmic with sheet resistance $R < 1 \text{ k}\Omega/\text{square}$ even when operated at $T < 1 \text{ K}$ as required for quantum computation, so doped features down to the single-nanometer scale can serve as qubit control wiring.²⁶ By contrast, host Si conductivity is frozen out, insulating APAM features from each other.²⁰

APAM device design utilizes doped feature sizes, shapes, and spatial separations to control key qubit properties, including Coulomb charging energy, tunnel and

Figure 4. (a) A schematic that shows the 3D layer structure of an atomic-precision advanced manufacturing two-qubit donor spin device. All qubit features (red) are dense 2D Si:P. (b) A scanning tunneling microscope image showing donor-doped qubit dots near a single-electron transistor reservoir. Reproduced from Kranz et al.³⁹ Figure 1, and licensed under Creative Commons 4.0 (<https://doi.org/10.1002/adma.202003361>).

exchange coupling, gate capacitive couplings, and electrical isolation.^{25–27} Most APAM devices have had coplanar doped features, as in Figure 1c, prepared on a (100)-vicinal surface utilizing one layer each of lithography, doping, and Si encapsulation. For a rough sense of scale, 10–40 nm interfeature distances are used for tunnel or exchange coupling, whereas distances > 50 nm electrically isolate structures (e.g., electrostatic gates) from each other. The isolation distance is balanced by a competing need to sustain capacitive coupling between features (e.g., dopants and gates). The electrical isolation pitches (> 50 nm) are incompatible with shorter two-qubit interaction length (< 40 nm), pointing to future in-plane crowding challenges.

APAM has proven to be an important scientific tool, with the potential for technological impact. There has been significant and accelerating progress over the past 10 years. Recent state-of-the-art developments include the following: (1) near

deterministic atomic control of doping in all three dimensions;^{28–30} (2) 3D devices with top gates aligned to buried structures;^{28,30} (3) alternative dopants – acceptors (B³¹, Al³²) and a donor (As³³); and (4) alternative resist chemistries (halogens) that take APAM into vacuum (> 10^{−8} torr) found in conventional Si foundries (see Fig. 3).³⁴

APAM quantum devices

APAM is the only known route to 3D atomistic control of dopant position and numbers. This exquisite control of dopant placement has led to important quantum electronics discoveries (e.g., exceptional electron spin relaxation times) ($T_{1e} = 30$ s) in devices and the first solid-state atomic two-qubit gates.^{21,22} With parallel learning from implant qubits ($T_{2e}^* \sim 0.27$ ms) in ²⁸Si, these discoveries are promising building blocks for quantum computation, but also expose critical challenges to scale up (e.g., 15–30 nm) interqubit pitches for robust, rapid, and two-qubit gates.¹⁰

Donor electron spin qubits

A two-qubit device is described in Figure 4. Spin qubits are hosted on *P* clusters (dots) of various sizes (1, 2, 3, ... *P*). APAM unlocks systematic studies with different dot sizes and spacings, leading to significant

recent discovery.^{35–40}

Dot size is determined to be valuable for separately addressing close-packed qubits at unique ESR frequencies owing to different hyperfine interaction strengths (97.5 MHz for 1*P* versus 262 MHz for 2*P*).²¹ Single-qubit ESR gates have been implemented,²¹ but gate fidelity (*F*) is not yet reported. Notably, ion-implanted single-qubit gates achieve *F* > 99.94% for both electron and nuclear spins.¹⁰

Watson et al. showed that spin relaxation time, T_{1e} , increases with qubit dot size yielding a record $T_{1e} = 30$ s for Si dot qubits using 2 and 3*P*.³⁵ Such long relaxation times aid high-fidelity readout. Strong reproducible tunnel coupling accessible via APAM leads to single-shot readout fidelity *F* > 99.7% with 62 ms readout time and *F* = 97% in rapid 1.5 μs readout.³⁷

Additionally, owing to precisely controllable strong exchange coupling, this type of device has uniquely enabled atomic singlet-triplet qubits³⁸ and two-qubit gates.²²

Figure 5. A schematic showing an atomic-precision advanced manufacturing (APAM) donor spin computer architecture.⁴² (a) Three layers of features are separated by ~40 nm. The middle layer contains qubits and single-electron transistor (SET) islands, and the top (bottom) layer contains source (drain) and control gate electrodes. The electrodes on the top layer are arrayed perpendicular to those on the bottom. (b) The SET islands (~13 × 13 nm²) each feed four P donors located ~15 nm from the SETs, which are reservoirs for spin initialization and readout, as in planar devices. (c) The electrodes are arranged such that a dopant is located in the center of the boundary formed by the S, D, G_A, and G_B electrodes. As shown, the S (D) electrodes alternate with the G_A (G_B) electrodes on the top (bottom) layer. (d) For initialization and readout of an individual qubit, the neighboring SET island is activated with the appropriate S/D pair (highlighted black), along with the highlighted G_A, G_B electrodes (yellow). Precise placement of the P qubits, as well as the other features, requires APAM techniques for fabrication.

The first two-qubit gate on atomic spins in Si

For two-qubit gates, native spin interactions include exchange and magnetic dipole coupling. Exchange naturally implements the so-called quantum SWAP^a gate, which is a useful primitive to construct other gates, and combined with one-qubit gates leads to a complete gate set for logic.¹⁰ The most straightforward variant is SWAP, which swaps qubit states. Halving the exchange interaction duration or strength yields a SWAP^{1/2} gate that creates two-qubit superpositions, e.g.: ($|\uparrow\downarrow\rangle \rightarrow \frac{1}{2}[(1+i)|\uparrow\downarrow\rangle + (1-i)|\downarrow\uparrow\rangle]$).

The first two-qubit gates on dopant spins were demonstrated at UNSW.²² An exchange-based SWAP^{1/2} gate was implemented on the device in Figure 4. Each qubit dot is initialized to a known spin state, then the exchange interaction is controllably increased using electrostatic gate voltages to push spin wavefunctions into closer overlap. By precisely tuning the overlap (exchange) strength and duration (here, 800 ps), a SWAP^{1/2} gate is achieved. The spin state of each qubit is then separately read out.

The overall two-qubit gate fidelity achieved with the device in Figure 4 is $F = 86.7 \pm 0.2$ percent. SWAP and SWAP²

gates were also compiled, but with diminishing fidelities limited by low-frequency electronic noise. Recent work indicates that fidelities > 99.9% adequate for larger-scale error-corrected quantum computation could be achieved by modestly increasing the speed (strength) of gate operations.³⁹

The control over dopant number and position that enabled this two-qubit gate is only accessible by APAM. First, repeatable two-qubit exchange gate interaction requires precise replicable distances between dots, which had been determined from significant work by the UNSW group. Second, qubits based on few donor dots (APAM only) mitigate extremely rapid Å-scale valley-orbit interference effects (Figure 2d), simplifying future scale-up.⁴⁰

Challenges and opportunities

The immediate path forward for dopant spin qubits, that incidentally aligns to APAM's key strengths, is to demonstrate high-fidelity (> 99%) two-qubit entangling gates with rigorous quantum tomography.⁴¹ Once few-qubit entanglement is mastered, dopant spins will be substantive building blocks for universal quantum computation, but the daunting multifaceted challenges of scale-up become acute. High-fidelity few-qubit APAM circuitry seems assured; therefore, we will highlight longer-term scale-up challenges and opportunities. There are three sets of problems: (1) finding architectures that expand the size and complexity of circuitry compatible with available two-qubit gate interactions; (2) developing APAM techniques to fabricate appropriate larger-scale circuits; and (3) needed materials developments (e.g., APAM-compatible ²⁸Si epitaxy).

Toward multiqubit architectures

Short interqubit distances (15–30 nm) forced by native spin physics point to difficulty for larger-scale donor spin circuitry owing to crowding of qubit control features. Some solutions to ease in-plane crowding, thereby making it possible to couple more qubits into circuits, are as follows: (1) reducing numbers of qubit control features, such as charge reservoirs and gates; (2) spacing-out to 3D multilayered structures; and (3) finding longer-range (>> 30 nm) gate interaction physics to space out qubits.

Toward multiqubit architectures

Figure 5 shows one proposed, possibly scalable, APAM architecture based on spin resonance one-qubit gates and

magnetic dipole coupling two-qubit gates.⁴² To ease 2D crowding, a 3D cross-bar scheme efficiently utilizes control lines spaced out vertically onto multiple planes separated by epitaxial Si. Fabricating this architecture will require significant APAM technique developments including nanometer-precision alignment between multiple layers, developing vertical tunneling devices (SETs), and increasing APAM lithography speed and yield to produce more than a few unit cells of the architecture in reasonable time. Recently, 3D multilayer APAM devices have achieved few-nanometer alignment accuracy between layers.²⁸

Scaling-up APAM techniques

For APAM to remain a valuable tool in the quantum scale-up era, methods must be developed to scale up, speed up, and broaden the technique. Some essential developments are (1) discovery of more accurate, precise, reliable, and rapid

parallel (versus serial) HDL; (2) increasing single P doping yield via greater control over incorporation chemistries; (3) epitaxial growth for isotopically enriched ^{28}Si ; and (4) extending APAM to new chemistries (e.g., acceptor dopants, for novel qubit schemes, to ease crowding and coupling challenges of donors).

STM HDL is a slow serial process requiring hours to fabricate small devices, see Figure 4. The speed limit is set by a required e-dose/area ($\sim 4 \text{ mC/cm}^2$) to stimulate H desorption at practical currents $\leq 100 \text{ nA}$, giving overall write speeds $\sim 10 \text{ nm}^2/\text{s}$. For true scale-up, the more important hurdle is to create massively parallel HDL methods. There have been a variety of proposals to overcome these limitations, including large arrays of microelectromechanical system (MEMS)-actuated tips to replace bulky piezo actuators, and STM tool development is an active R&D topic.^{43–45}

A less obvious challenge to scaling is that P incorporation yield is < 100 percent. Due to the complex, nonequilibrium, and inherently stochastic surface chemical processes involved, the highest reported single-P yields are ~ 0.7 .^{46,47} This yield will cause significant difficulties, since the yield will be 0.7^N for circuits with N single-atom qubits.

For example, for the $N=4$ unit cells in the architecture in Figure 5b, the yield would be $0.7^4 \sim 24\%$ for perfect $N=4$ P cells. It is not entirely certain that yield ~ 0.7 is a fundamental limit, and more work is required to understand how to improve single-P yield.^{47,48}

Figure 6 overviews a simplified chemical pathway illustrating the difficulty. In particular, as shown in Figure 6a, to place a single-P atom, HDL templates consisting of six exposed Si atoms are ideal, into which up to three precursor molecules adsorb. During a subsequent incorporation anneal, some doping precursor (PH_3) will desorb, leading to the observed ~ 0.7 single-P yield. Larger patterns (8–12 sites) increase the probability toward one, with a new difficulty that two or more P atoms may also be incorporated, completely changing the electronic structure of the qubit. Figure 6b shows the HDL and doping outcomes of a 25-qubit sample array targeting single-donor placement, with 1P yield of 13/25, illustrating challenges for

both HDL (errors, disorder) and subsequent doping (varying single- P yield due to 0, 1, 2, ..., P incorporation).

Recently, various doping temperature and pressure regimes have been proposed to increase P incorporation to 100 percent.^{47,48} Another proposed solution is to increase P yield and accuracy by driving dissociation deterministically (Figure 6c). By using the STM tip to remove H atoms from the adsorbed PH_3 , the pathway to re-desorption is blocked.⁴⁹ Thus, an alternative process would be, as shown Figure 6c, to adsorb a single PH_3 in a two-atom pattern, remove H from the PH_3 and surrounding Si, and then do an incorporation anneal.

Scaling-up and implementing error correction will require optimally long spin dephasing times (T_2^*), requiring a technique for isotopically enriched ^{28}Si epitaxy to encapsulate APAM devices. While ion-implanted qubits are fabricated on enriched ^{28}Si layers prepared by chemical vapor deposition (CVD), APAM devices require encapsulation to bury the P atoms. However, low-temperature ($<300^\circ\text{C}$) processes are required to limit disordering via dopant diffusion. Such low temperatures are not ordinarily compatible with CVD processes, but MBE has been effective at these temperatures.^{50,51} One promising solution is to use mass filtered molecular beam epitaxy, which can yield $>99.999\%$ ^{28}Si epitaxy at APAM-compatible temperatures,⁵⁰ although low-temperature CVD encapsulation has not been explored.

Owing to crowding difficulties facing spin circuitry, and APAM architectures, multiqubit scale-up is likely to be more straightforward using longer-range interactions than donor spin–spin exchange.⁵² Donor interactions using sophisticated arrangements such as exchange-coupled spin chains or interface MOS dots (e.g., for spin coherent transport by adiabatic passage)⁵³ appear to add complexity with minimal reduction in circuitry density. Coherent coupling mediated by circuit and cavity quantum electrodynamics (cQED) methods is possible, but likely to be challenging due to weak cavity-spin magnetic coupling.¹⁰

A novel approach to mitigate coupling and crowding problems is acceptor hole spin qubits. By virtue of Rashba-like spin–orbit coupling inherent to holes near interfaces, multiqubit electric dipolar coupling greater than the 100-nm-range and all-electrical qubit control can be achieved, while preserving other key qubit properties (e.g., long T_2^*).^{54,55} Furthermore, acceptor holes can couple efficiently via cQED methods using both photons and phonons, for qubit interaction across chip-scale distances.⁵⁵ Although APAM is not necessarily urgent for investigating acceptor qubits, it could plausibly serve as a means to make systematically reproducible structures, highlighting the important role of APAM for scientific discovery and prototyping. Recently, significant work aims to expand APAM to achieve acceptor doping comparable to current donor methods.^{31,32}

Summary

Silicon-based quantum computation with dopant spins is a promising platform for scale-up and is currently maturing toward high-fidelity building blocks required for

fault-tolerant quantum computation. The forefront of dopant spin qubit research, in general for both APAM and ion implant, is high-fidelity two-qubit gates. Development of two-qubit systems has defined some challenges for spin architectures and APAM techniques. For the foreseeable future, the main role for APAM is to provide researchers access to precisely controlled, pristine, reproducible, and small-scale circuitry to investigate the physics of solid-state atomic quantum computation. Regardless of whether APAM itself scales or plays a direct role in realizing any larger future quantum computation technologies, APAM significantly augments researchers' ability to create and learn at atomic, and inherently quantum, scales via deterministic control over dopant numbers and locations at scales much less than characteristic lengths for quantum interactions (~ 3 nm/decade for exchange and tunneling). Subsequently, APAM lessons can be extended to quantum devices fabricated using scalable Si foundry craft.

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Conflict of interest

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

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