Towards the fabrication of phosphorus qubits for a silicon quantum computer

J. L. O’Brien,1,2 S. R. Schofield,1,2 M. Y. Simmons,1,2 R. G. Clark,1,2 A. S. Dzurak,1,3 N. J. Curson,1,2 B. E. Kane,4 N. S. McAlpine,4 M. E. Hawley,6 G. W. Brown6

1 Centre for Quantum Computer Technology, University of New South Wales, Sydney 2052, Australia
2 School of Physics, University of New South Wales, Sydney 2052, Australia
3 School of Electrical Engineering & Telecommunications, University of New South Wales, Sydney 2052, Australia
4 Laboratory for Physical Sciences, University of Maryland, College Park, MD 20740, USA
5 Los Alamos National Laboratory, Los Alamos, NM 87545, USA

(October 29, 2001)

The quest to build a quantum computer has been inspired by the recognition of the formidable computational power such a device could offer. In particular silicon-based proposals, using the nuclear or electron spin of dopants as qubits, are attractive due to the long spin relaxation times involved, their scalability, and the ease of integration with existing silicon technology. Fabrication of such devices however requires atomic scale manipulation - an immense technological challenge. We demonstrate that it is possible to fabricate an atomically-precise linear array of single phosphorus bearing molecules on a silicon surface with the required dimensions for the fabrication of a silicon-based quantum computer. We also discuss strategies for the encapsulation of these phosphorus atoms by subsequent silicon crystal growth.

(To appear in Phys. Rev. B Rapid Comm.)

A quantum bit (or qubit) is a two level quantum system that is the building block of a quantum computer. To date the most advanced realisations of a quantum computer are qubit ion trap computers. To date the most advanced realisations of a quantum computer are qubit ion trap computers. To date the most advanced realisations of a quantum computer are qubit ion trap computers. To date the most advanced realisations of a quantum computer are qubit ion trap computers. To date the most advanced realisations of a quantum computer are qubit ion trap computers.

The requirements for this quantum computer design are very stringent. In order to undertake high resolution lithography the silicon surface must be atomically flat with a low defect density to allow the formation of a near perfect resist layer, where one hydrogen atom bonds to each surface silicon atom. The ability to then desorb individual hydrogen atoms requires a sharp, large cone angle tungsten tip in order to form <1 nm desorption sites. These sites are subsequently exposed to high purity...
FIG. 1. A schematic of the process to fabricate the Kane architecture. (a) Detail of the Kane quantum computer architecture taken from Ref. 7, showing two phosphorus qubits in a linear array, incorporated into isotopically-pure $^{28}\text{Si}$ and isolated from surface metal A and J gates by an insulating barrier. (b) A process to fabricate an array of phosphorus qubits in silicon. A low defect density Si(001)2x1 surface is passivated with a monolayer of hydrogen. An STM tip is used to selectively desorb hydrogen, exposing silicon on an atomic scale permitting only one phosphine molecule to adsorb at each of the required sites. Low temperature silicon overgrowth encapsulates the phosphorus.

phosphine gas for the required phosphorus atom placement (Fig. 1b). We demonstrate each of these steps below. Throughout this process particular attention must be made to avoid the introduction of any spin or charge impurities that would be fatal to the operation of the quantum computer.

The final fabrication step outlined in Fig. 1b which is not the subject of this paper, but presents a significant challenge, is to encapsulate the phosphorus qubits in a crystalline lattice of isotopically-pure silicon-$^{28}\text{Si}$. The main difficulty in this step is to ensure the phosphorus atoms incorporate into the silicon crystal and remain in their ordered atomic array. The most direct route to achieving this is to desorb the hydrogen resist from the surface by heating to $\sim 700\, \text{K}$ followed by epitaxial silicon growth over the phosphorus array. A possible concern with heating the surface is the potential to induce lateral surface diffusion of the phosphorus atoms in the array. However, it is known that the phosphorus atom in the PH$_2$ molecule, with a single bond to the silicon, incorporates into the silicon surface with a more stable threefold coordination geometry over the lower temperature range of 550-650 K. We can avoid heating the surface at all during hydrogen desorption by either direct optical excitation at $\lambda = 157\, \text{nm}$ or electron bombardment. An alternative room temperature approach that will help to incorporate the phosphorus into the silicon lattice without significant diffusion involves photo-induced excitation of the attached PH$_2$ molecule to increase its chemical activity with the surface. Finally it is also possible to leave the hydrogen resist and grow epitaxial silicon directly on the mono-hydride surface. In all above cases 10-30 Å of epitaxial silicon can be grown at low temperatures, down to room temperature, to encapsulate the phosphorus, followed by elevated temperature growth at $\sim 500\, \text{K}$ to maintain crystallinity. We will return to the issue of phosphorus incorporation in our final discussion.

The first step in fabricating a phosphorus qubit array is to prepare a clean, defect free silicon surface. Fig. 2a demonstrates an optimally-prepared Si(001)2x1 surface consisting of rows of $\sigma$-bonded silicon-silicon dimers. By heating the surface to 1200 °C in a variable-temperature ultra-high vacuum STM system and performing a controlled cool-down we have achieved large defect free regions. The “bean” shaped protrusions in this filled state image correspond to the charge overlap of the electrons in the dangling bonds on each silicon surface atom giving rise to a weak $\pi$-bond. These dangling bonds make the surface reactive, allowing the subsequent adsorption of species such as hydrogen and phosphine.

The adsorption of atomic hydrogen on the Si(001)2x1 surface can occur in three ways depending on the surface temperature, forming either a mono-, di- or tri-hydride. An atomic hydrogen source consisting of a heated tungsten filament and water-cooled heat shroud has been used for this work. Fig. 2b shows an image taken at room temperature of a low defect density Si(001)2x1 surface after exposure to high purity atomic hydrogen at 600
FIG. 3. Bonding structure of phosphine on Si(001)2x1. (a) Schematic of the c(4×2) structure of PH$_3$ bonded to the Si(001)2x1 surface, where red dimers indicate PH$_3$ bonding sites. (b) An STM image of this structure, acquired at a sample bias of -3.0 V and tunnelling current 0.2 nA. (c) STM image of three hydrogen desorption sites on a mono-hydride Si(001)2x1:H surface. The highlighted regions in (a) and (c) indicate that only one phosphine molecule can adsorb at each desorption site.

K. It can clearly be seen that a near uniform coverage of the mono-hydride phase has occurred, where one hydrogen atom bonds to each silicon atom. Sites of dihydride, where two hydrogen atoms bond to each silicon, and tri-hydride, a mixture of the other two phases, are also indicated. We have found that all three phases passivate the surface and act as an effective resist during subsequent exposure to phosphine. Comparison of current-voltage spectroscopy before and after hydrogen dosing (not shown) confirms the existence of a hydrogen passivated surface, with the silicon π*-antibonding peak evolving into the silicon-hydrogen antibonding peak after hydrogenation.

An STM tip is then used to desorb hydrogen from the surface by application of a controlled voltage pulse between the tip and sample. Optimisation of the geometry of an oxide-free tungsten tip and controlled voltage pulses (sample bias ~6 V and tunnelling current ~1 nA for ~1 ms) makes atomic resolution desorption possible. In order to allow the adsorption of one phosphine molecule, and therefore only one phosphorus atom, it is necessary to desorb an area that exposes less than or equal to two silicon dimers as shown schematically in Fig. 3a. This is because phosphine bonds to the Si(001)2x1 surface with a c(4×2) surface periodicity as demonstrated in Fig. 3b where we have dosed a clean Si(001)2x1 surface with phosphine. The STM image in Fig. 3c shows three < 1 nm diameter hydrogen desorption sites in a row with a pitch of ~4 nm on a hydrogen terminated Si(001)2x1 surface. This image, with such a close spacing between sites, highlights the atomic resolution desorption achieved. The distance between sites can easily be increased to the required qubit spacing of 20 nm, and we have performed controlled lithography of single desorption sites in a line > 100 nm in length. The desorption sites in Fig. 3c appear as bright protrusions as a result of the extension of electron density out of the surface due to the silicon dimer surface states of the exposed silicon dangling bonds.

FIG. 4. Demonstration of single PH$_3$ adsorption through a STM patterned hydrogen resist. STM images of two desorption sites before (a) and after (b) phosphine dosing. The corresponding line profiles (c and d) show a characteristic height increase of ~0.05 nm. Three desorption sites before (e) and after (f) phosphine dosing and corresponding line profiles (g and h). All images were acquired at a sample bias of -1.8 V and tunnelling current of 0.4 nA.
The absence of available dangling bond sites nearby for the re-adsorption of the dissociated Si(001)₂×₁ surface atoms in the centre are surface atoms and the others are sub-surface atoms. In order to obtain high purity phosphine gas delivery, the PH₃ micro-dosing system and its connection to the UHV STM employed internally electro-polished gas lines assembled in a clean-room environment. Mass spectra taken in the chamber during the exposure at a pressure of 10⁻⁸ mbar reveal no significant increase in the partial pressure of any other species. The sticking coefficient of phosphine on the clean silicon surface is 1.9. Dissociation of singly-bonded PH₃ to threefold coordinate P + 2H can proceed above 500 K, provided there are suitable dangling bond sites that overcome competition with the recombinative desorption of PH₃. An alternative option is to dose the hydrogen-patterned surface with phosphine gas at elevated temperatures (∼550 K) to directly induce phosphorus incorporation into the first atomic layer. We have independently performed elevated temperature studies which indicate that at these temperatures, the silicon surface is stable.

We have demonstrated the effectiveness of a hydrogen resist as a barrier to phosphine adsorption and used STM lithography for the controlled placement of single phosphorus bearing molecules on a silicon surface - central to the construction of a silicon based quantum computer. This process, shown for closely spaced controlled doping, demonstrates the achievability of more widely-spaced (∼20 nm), precisely positioned phosphorus qubit arrays over large areas. Whilst it is not yet possible to guarantee the subsequent overgrowth steps required for the Kane architecture, this letter demonstrates a significant advance in the solid-state qubit fabrication process and bodes well for the realisation of a scalable silicon quantum computer. Finally, it is important to note that the fabrication strategy demonstrated here is also directly applicable to other silicon based quantum computer architectures.

We would like to thank I. Andrienko, T. Kamins, G. J. Milburn, S. Prawer, C. Thirstrup and S. Williams for useful discussions. This work is funded by the Australian Research Council Special Research Centre scheme, the Australian Government, the US National Security Agency, and the US Advanced Research and Development Activity.
A larger height in the line profile of the dimer rows is observed after PH$_3$ dosing. Such a difference can frequently occur due to minor changes in imaging conditions between scans, which results in the STM tip extending further into the gap between dimer rows. However, the height difference due to PH$_3$ adsorption is measured from the top of the dimer rows to the top of the protrusion and is not therefore affected by this.

Zhi-Heng Loh, H. C. Kang, J. Chem. Phys. **112**, 2444 (2000) and references therein.

J. E. Northrup Phys. Rev. B **51**, 2218 (1995).