Quantum-information processing with ferroelectrically coupled quantum dots

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A proposal is described to construct a quantum-information processor using ferroelectrically coupled Ge/Si quantum dots. The spin of single electrons form the fundamental qubits. Small-diameter (<10 nm) Ge quantum dots are optically excited to create spin-polarized electrons in Si. The static polarization of an epitaxial ferroelectric thin film confines electrons laterally in the semiconductor; spin interactions between nearest-neighbor electrons are mediated by the nonlinear process of optical rectification. Single-qubit operations are achieved through ''*g*-factor engineering'' in the Ge/Si structures; spin-spin interactions occur through Heisenberg exchange, controlled by ferroelectric gates. A method for reading out the final state, while required for quantum computing, is not described; approaches involving single-electron transistors may prove fruitful in satisfying this requirement.

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I. INTRODUCTION

Quantum-information processing is a central component for quantum computation [1], cryptography [2], and secure communication over large distances [3]. A quantum-information processor must generate or accept an entangled quantum state as input, perform controlled operations on that state, and produce an output for measurement or further processing. The requirements for implementing such a system are stringent, and the technical challenges great. The ultimate form that a successfully engineered quantum-information processor takes will determine, in large part, the architecture of technologies that utilize these new capabilities.

Groundbreaking experiments by Kikkawa *et al.* have given hope that semiconductor-based architectures that utilize the spin of electrons will enable these requirements to be met. Experiments in direct-band semiconductors such as GaAs have shown that electron spins exhibit a long-lived coherence that persists to room temperature [4] and is robust against diffusion [5] and transport across interfaces [6]. However, electronic spin lifetimes in III-V and II-VI systems are limited by spin-orbit and hyperfine coupling. Si is an attractive material for spin-based quantum computation [7] because of weak spin-orbit coupling and because Si is primarily composed of isotopes with nuclear spin zero; consequently, decoherence times of the electron spin can be exceedingly long [8].

A number of solid-state spin-based approaches to quantum information processing have been proposed in which localized states are manipulated through externally applied gate voltages [7,9,10]. Still, several hurdles remain. The control gates, especially those that mediate interactions between electron spins, must be modulated extremely rapidly and accurately, near or beyond the limit of what is achievable with current technology. Because the separation between bound states must be comparable to the Bohr radius for the electron, extremely high-resolution lithography is also required in most cases.

Here I describe an approach to quantum information processing that uses the static and dynamic polarization of a ferroelectric thin film to control electron spin interactions in Si. Two properties of ferroelectric materials are involved: (1) the use of static ferroelectric domains that confine laterally electrons in an adjacent semiconductor, and (2) a large nonlinear response (optical rectification) that enables terahertz polarizations to develop and influence nearby electronic states.

II. REQUIREMENTS FOR QUANTUM COMPUTING

The requirements for quantum computing are exceedingly stringent, and have been delineated by a number of authors [1,11,12]. Here I follow the exposition of DiVincenzo [11]. who outlines five requirements, summarized in the first column of Table I. The second column of Table I summarizes how these requirements will be met using ferroelectric/semiconductor heterostructures. The remainder of this paper will expand on the proposal as outlined in Table I.

Requirement $(R1)$ states that there must be a scalable architecture with a well-defined two-level system. The spin of an electron (or $I=\frac{1}{2}$ nuclear moment) forms a natural two-level system that is only weakly coupled to other degrees of freedom. In recent years, there has been a great deal of interest in exploring and exploiting the spin degree of freedom in electronic systems. With conventional electronics, the spin of the electron is essentially irrelevant; the importance of the spin degree of freedom is epitomized in the way that the electron wave function is commonly expressed [see Eq. (1a)]. By contrast, with ''spintronics,'' it is the spin degree of freedom that contains the relevant quantum information; the spatial part of the wave function provides a convenient ''handle'' by which spins are transported and made to interact [see Eq. (1b)]:

$$\Psi_e = \psi(\vec{r})\chi_s, \tag{1a}$$

$$\Psi_e = \chi_s\psi(\vec{r}). \tag{1b}$$

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TABLE I. Left, Di Vincenzo's summary of the requirements for quantum computing [34]. Right, summary of proposed method for meeting these requirements.

	Requirement	Proposed implementation
$(R1)$	A scalable physical system with well-characterized qubits	Electron spins in Si
$(R2)$	The ability to initialize the state of the qubits to a simple fiducial state, such as $\|000\cdots0\rangle$	Optical spin injection into Si using quasidirect gap Ge quantum dots
$(R3)$	Long relevant decoherence times, much longer than the gate operation time	Long spin lifetimes in Si: fast (2-qubit) gate operation times using ferroelectric gates
$(R4)$	A ''universal'' set of quantum gates	*g*-factor engineering and Heisenberg exchange using optical rectification in ferroelectric
$(R5)$	The ability to measure specific qubits	Single electron transistors (nonoptical)

Requirement $(R2)$ is important for ''booting'' the quantum computer. The problem of initializing a collection of spins in semiconductors to a specific repeatable state is nontrivial. One way to achieve this result is to place them in a magnetic field at low temperature. To achieve sufficiently high polarization in bulk Si using a magnetic field strength $B=2$ T, very low temperatures ($T\sim100$ mK) are required. Because the equilibrium spin polarization is a function of B/T, larger fields could be used at higher temperatures. However, smaller fields are preferable because many decoherence mechanisms scale as higher powers of B [13]. Optical pumping of spin-polarized carriers is a relatively straightforward method for producing spin-polarized electrons in direct-gap semiconductors such as GaAs [14]. Unfortunately, all-optical approaches are not simply extended to indirect-gap semiconductors such as Si and Ge. Direct optical transitions can be engineered in Si by creating small Ge/Si quantum dots;[15–19] these quantum dots may provide a method for meeting requirement $(R2)$.

Requirement $(R3)$ states that there must be a separation of time scales (by at least 10^5) that characterize the gate time and the decoherence time in the system. Both time scales depend on the physical system, as well as the way in which the subsequent requirement $(R4)$ will be satisfied. The weak spin-orbit coupling in Si ($|g-2|<3\times10^{-3}$) leads to long transverse decoherence times for donor electrons in Si [$T_2=5\times10^{-4}$ s for dilute *p* donors in isotopically enriched Si (Ref. [8])]. Strain and the introduction of other materials such as Ge are expected to lead to reduced values for T_2; nevertheless, these times are still long compared to the best values reported for GaAs [4].

Requirement $(R4)$ relates to the central processing unit of the quantum computer—the quantum logic gates that process quantum information. A minimal set of one-qubit and two-qubit gates is required for quantum computation. There are two ways in which ferroelectric materials will be used in satisfying $(R4)$. The first has to do with controlling the static interactions between electrons through ferroelectric domain patterning. Ferroelectric materials are useful ''hands'' that can manipulate the charge degree of freedom with the necessary high spatial and temporal resolutions required for quantum-information processing. Recent advances in materials by McKee, Walker, and Chisholm [20] have resulted in high-quality *crystalline* interfaces between oxide ferroelectrics and semiconductors. For example, bulk $BaTiO_3$ has a spontaneous polarization $P_s=26\,\mu C/cm^2$ at room temperature, corresponding to a sheet density $\sigma=\pm1.6\times10^{14}$ cm^{-2}. The field effect at the ferroelectric/semiconductor interface is so strong that quantum confinement effects should be achievable with suitably small domain patterning. The second property of ferroelectric materials is their nonlinear response to electromagnetic fields. Optical rectification, a manifestation of the linear electro-optic effect, allows an optical field to be converted into a terahertz polarization that persists as long as the optical pulse is present. Terahertz ferroelectric switches may allow the necessary control over electron spin interactions required to create exchange gates.

Requirement $(R5)$ delineates the requirements for final-state measurement. Optical methods such as Faraday rotation have a low ''quantum efficiency,'' and cannot measure single-quantum events with high fidelity. It is most likely that electronic methods, perhaps involving single-electron transistors [21], will provide the necessary means for satisfying this requirement. It could be argued that a quantum-information processor need only satisfy requirements $(R1,3,4)$, since it involves neither state preparation nor measurement.

III. MATERIAL SYSTEMS

The viability of the proposed approach to quantum-information processing depends critically on the material systems and their properties. Below I discuss two important subsystems and their relevant properties.

A. Ge/Si quantum dots

Bulk Si and Ge are indirect-gap semiconductors that absorb or emit photons only with the assistance of a phonon to conserve momentum. As a consequence, the optical selection rules that enable spin-polarized carriers to be created in direct-band semiconductors such as GaAs are absent from such materials [22]. In order to initialize the state of a quantum computer [requirement $(R2)$], it will be important to engineer direct optical transitions in Si using Ge quantum dots. These quantum dots can also be used to localize the electrons.

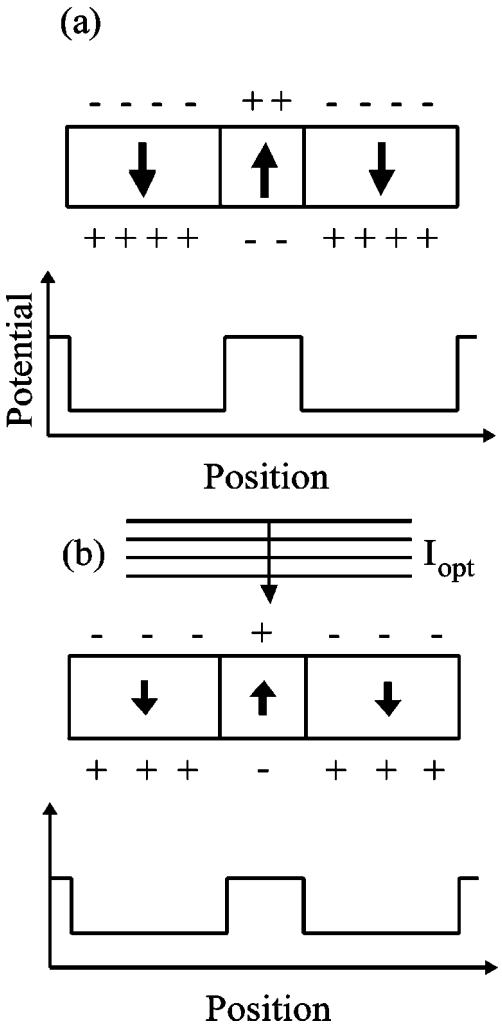

FIG. 1. Schematic of ferroelectric domains and optical rectification. (a) $I_{opt}=0$. (b) $I_{opt}>0$. The magnitude of the ferroelectric polarization decreases upon illumination.

One way to convert an indirect semiconductor into a direct one is to form a superlattice (i.e., Si and Ge) [23]. In doing so, the size of the Brillouin zone is reduced and a portion of the conduction-band minimum is folded onto the Γ point, resulting in a quasidirect gap system. For semiconductor quantum dots, a momentum-space representation is not appropriate, but if the envelope wave functions of confined carriers have a sizeable Fourier component at the wave vector corresponding to the direct gap transition, then direct optical transitions become allowed. Takagahara and Takeda [24] have calculated radiative rates for Ge quantum dots, showing an indirect-direct crossover near a diameter $d \sim 10\,\mathrm{nm}$. Naturally formed Si/Ge quantum dots grown by self-assembly using the Stranski-Krastanov mode have a range of diameters that start at approximately 20 nm and increase to as large as 50–100 nm. These quantum dots are *not useful* for quantum computing applications, because they are too large to have appreciable direct optical transitions. In order to grow quantum dots with smaller sizes, several groups have deposited a nucleation layer, using either semiconducting (C) (Refs. [15] and [17]) or *n*-type (Sb) (Ref. [19]) materials; both have been demonstrated to yield quantum dots with strong luminescence and signatures of direct optical transitions [18,19]. It has also been shown [15] that conditions exist in which quantum dots will grow spontaneously only when carbon is present. The small diameter and bright luminescence of these quantum dots are encouraging for optical control over the initial-spin state of the electrons.

In order to satisfy $(R2)$, it will be important to create only a single electron-hole pair at each quantum dot. A simple method for doing this involves pulsed resonant excitation of the single-exciton state. After one exciton is created, the Coulomb energy associated with the bound hole will inhibit the absorption of a second photon. A procedure for exploiting this effect to produce a current standard was first discussed by Kouwenhoven [25]. The Coulomb energy as a function of quantum-dot diameter d was explored by Yakimov *et al.* [26]. By monitoring the temperature dependence of the hole conductivity in Ge/Si[100] quantum dots as a function of size, and extracted effective Coulomb charging energies E_C as a function of dot diameter. For the smallest size studied ($d=12\,\mathrm{nm}$), they found $E_C=16\,\mathrm{meV}$, scaling as $1/d$. Transform-limited optical pulses generated by commercially available ultrafast laser sources should therefore satisfy the criteria needed for self-limiting generation of photoelectrons.

B. Ferroelectric thin films

Below, I review several relevant properties of ferroelectric materials and provide numerical estimates of these effects for reasonable parameter values.

1. Ferroelectric domain patterning

The ability to pattern ferroelectric domains forms a critical part of the proposed system. In particular, it is important to be able to write domain patterns on length scales comparable to the extent of the electron wave function (diameter $\sim 100\,\text{Å}$). Antiparallel (180°) domain walls in perovskite ferroelectrics such as $BaTiO_3$ are exceptionally thin, and are comparable in size to the unit cell (5–20 Å) [27]. In thin films, it is possible to use conducting atomic force microscope (AFM) tips to both write and read ferroelectric domains [28]. The same AFM tip can also be used to sense the ferroelectric polarization direction through the inverse piezoelectric effect [28]. However, the signal becomes quite small for ultrathin ferroelectric films ($\sim$100 Å), making direct detection of induced domain states difficult (Fig. 1). It is quite plausible that the domains that are actually produced in this manner are smaller than what can be measured using AFM techniques alone.

The polarization at the surface of the ferroelectric can have a significant influence on an epitaxially grown semiconductor. Recent results from McKee, Walker, and Chisholm [20] and McKee and Walker [29] show that lattice matched ferroelectric thin films, grown epitaxially by molecular beam epitaxy with a *crystalline* interface can produce exceptionally large ferroelectric field effects [30]. The ability to write ferroelectric nanodomains, along with the ferroelectric field effect, forms an important element in the proposed quantum-

information processing architecture.

2. Optical rectification

The induced polarization P in a nonlinear material is conventionally expanded in powers of the electric field strength E,

$$P_i = \chi_{ij}^{(1)} E_j + \chi_{ijk}^{(2)} E_j E_k + \cdots \equiv P_i^{(1)} + P_i^{(2)} + \cdots . \quad (2)$$

The second-order contribution leads to second harmonic generation, difference frequency generation (DFG), and optical rectification. Using the complex representation $\tilde{E}(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + \text{c.c.}$, one obtains for the case of DFG:

$$P_i^{(2)}(\omega_1 - \omega_2) = 2\chi_{ijk}^{(2)} E_{1j} E_{2k}^{*} . \quad (3)$$

For the case of a femtosecond pulse, the various Fourier contributions to DFG interfere to give a temporal profile for the nonlinear polarization that is proportional to the optical intensity I_{opt}:

$$P^{(2)}(t) = \frac{r n^3}{2c} I_{\text{opt}}(t), \quad (4)$$

where r is the electro-optic coefficient (indices suppressed for simplicity), n is the refractive index, and c is the speed of light. The numerical value of the maximum value for the polarization is expressed in terms of quantities relevant to $BaTiO_3$ and accessible in the laboratory:

$$P_{\max}^{(2)} = (6.29\times 10^{-2}\ \text{cm}^2/\mu\text{C}) \left(\frac{I_{\text{avg}}}{10\ \text{mw}}\right) \left(\frac{D}{1\ \mu\text{m}}\right)^2 \left(\frac{76\ \text{MHz}}{\Omega}\right) \times \left(\frac{r}{1.95\times 10^{-11}\ \text{m/V}}\right) \left(\frac{\tau_{\text{opt}}}{100\ \text{fs}}\right) \left(\frac{n}{2.45}\right)^3 , \quad (5)$$

where D is the laser spot diameter, Ω is the laser repetition rate, r is the electro-optic coefficient, τ_{opt} is the pulse width, and n is the refractive index. Because the nonlinear polarization arises from a nonresonant process below the optical gap of the semiconductor (and ferroelectric), it is possible to use large power densities without concern for photogeneration of carriers, heating, etc. For $BaTiO_3$, using a diffraction-limited spot with 10 mW average power, one obtains the following nonlinear polarization for $BaTiO_3$: $P_{\max}^{(2)} = 6.29 \times 10^{-2}\ \mu\text{C/cm}^2 = 3.93\times 10^{11} e^-/\text{cm}^2$. Because ferroelectric domains can be patterned with high precision, and because the sign of the electro-optic coefficient depends on the polarization direction, the spatial extent of the optically induced electric fields is *not* limited by diffraction. That is, the nonlinear polarization may be used to actuate a gate separating two electrons using optical fields that extend over much larger length scales.

Generation of propagating terahertz electromagnetic waves using optical rectification was achieved by Nahata and Heinz, who bonded a $LiTaO_3$ superstate to a coplanar microstripline [31]. Using modest energy densities, they measured significant voltages (>0.7 mV) 350 μm from the source. Both pulse broadening and the effects of bonding cause lowering of the effect. In epitaxial films, the separation between ferroelectric and semiconductor is measured in angstroms—hence, the effects are expected to be significantly larger for epitaxial ferroelectrics on semiconductors.

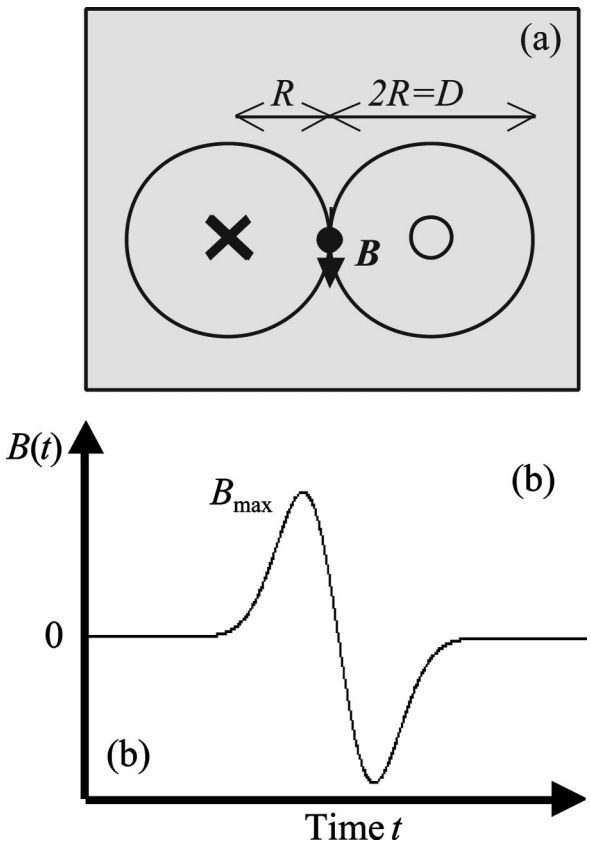

FIG. 2. (a) Geometry (top view) for estimating the magnetic field strength due to ferroelectric displacement currents. At left, a circular region is poled inward; tangent at right, a circular region poled outward. (b) Schematic of temporal profile for magnetic field induced by ferroelectric displacement currents.

3. Local magnetic fields

The transient displacement currents from ferroelectric domains will induce transient magnetic fields. While the exact profile depends on the details of the domain pattern, the overall scale can be estimated by considering a simple geometry, in which a circular cylinder of radius R sustains a displacement current density of magnitude $P_{\max}^{(2)}/\tau_{\text{opt}}$, as shown in Fig. 2. The maximum field strength is given by Eq. (6). For reasonable parameters, the peak magnetic field can be quite large; however, the temporal profile, which is proportional to the time derivative of the optical intensity $I_{\text{opt}}(t)$, limits its use for electron spin resonance. The combination of electric and magnetic fields may lead to useful geometric phases (i.e., Berry's phase) that can be exploited in single-spin phase shifts; however, significantly more detailed modeling of the physical system is required to assess the strength and usefulness of these effects. As a source of

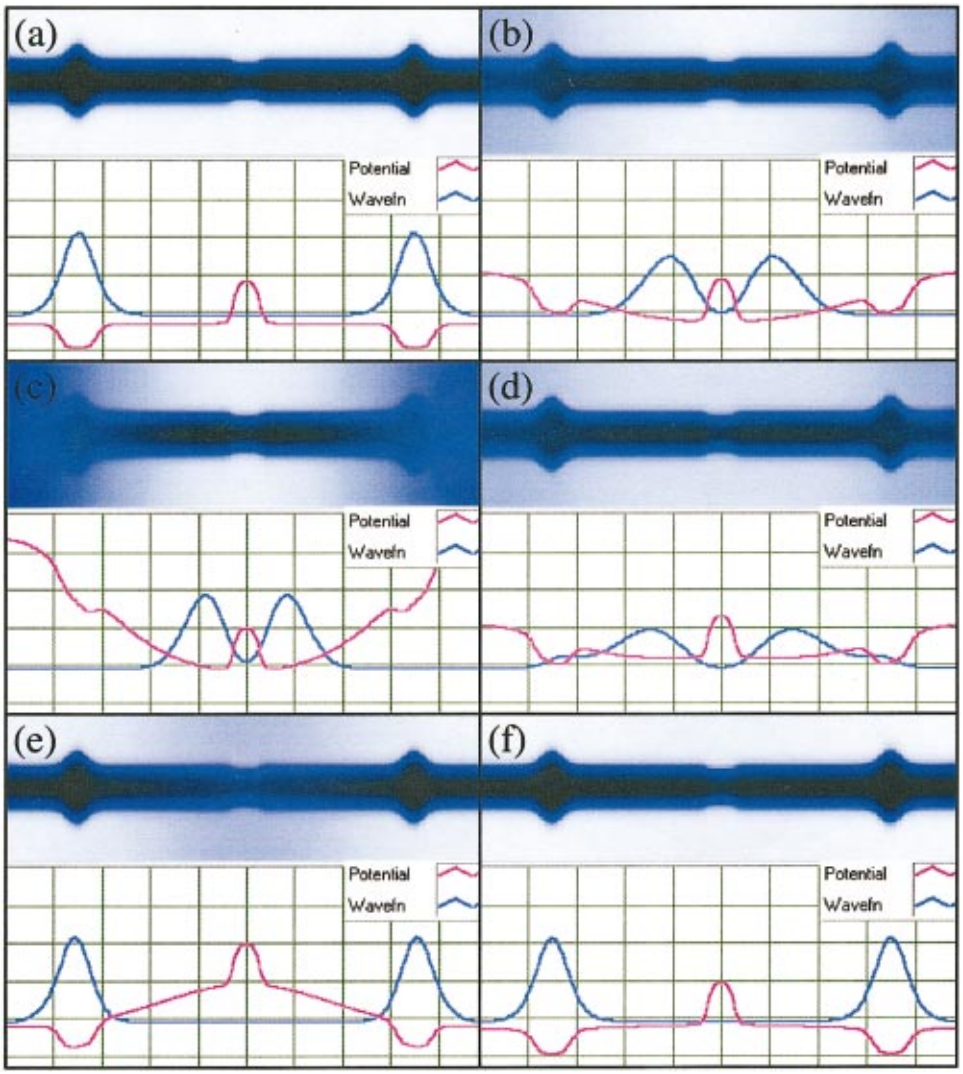

FIG. 3. (Color) Quantum swap operation. Electrons are guided along a ferroelectrically defined nanowire (top), with a tunneling ''kink'' at the center. Bottom graphs show 1D effective potential along the nanowire and corresponding ground state wave functions. Optical rectification of a femtosecond pulse generates a transient coupling between quantum dots. (a) $t=-200$ fs. (b) $t=-100$ fs. (c) $t=0$. Ionized electrons undergo controlled exchange. (d) $t=150$ fs. Coupling is turned off. (e) $t=450$ fs. Weaker second pulse centered on tunneling barrier returns electrons to their confined states. (f) $t=600$ fs. Controlled exchange operation is complete.

decoherence (systematic phase errors), it will be important to understand these contributions.

$$B_{\max}=\frac{\mu_0 R P^{(2)}_{\max}}{\tau_{\rm opt}} = (39.6\,\mathrm{G})\left(\frac{R}{0.5\,\mu\mathrm{m}}\right)\left(\frac{P^{(2)}_{\max}}{6.29\times10^{-2}\,\mathrm{cm}^2/\mu\mathrm{C}}\right)\left(\frac{100\,\mathrm{fs}}{\tau_{\rm opt}}\right). \tag{6}$$

In order to perform arbitrary quantum-logic operations, a ''universal'' set of gates is required [32]. Until recently, most electron-spin-based proposals worked to identify mechanisms that will enable both single-spin rotations and nearest-neighbor exchange interactions, which together form a universal set. However, single-spin operations are slow compared to exchange operations. Recent theoretical developments by Bacon and co-workers [33,34] have shown that it is possible to use decoherence-free subspaces of multispin registers as composite qubits, and that effective one-qubit and two-qubit interactions can be implemented using only (Heisenberg) exchange gates. There are two clear advantages of this approach to quantum computation. First, there is significant overhead involved in developing a quantum gate; reducing the set of required gates from two to one greatly reduces the burden on the experimentalist. Second, the exchange interaction is in principle very fast, if it can be controlled with sufficient precision. Using spatiotemporal pulse shaping methods, it should be possible to control the spacing and timing of these terahertz pulses and create the fundamental gate operations required for quantum-information processing.

IV. QUANTUM-LOGIC OPERATIONS

A. The quantum swap

At the heart of the quantum-information processor is a mechanism for controlling spin exchange between neighboring electrons. This interaction is achieved through the combination of static and terahertz fields of an epitaxial ferroelectric thin film. Ferroelectrically defined nanowires ensure that the electrons reach the tunneling barrier without ''wandering.'' Figure 3 depicts the transient potential induced by

optical rectification, and the induced exchange interaction through a ferroelectrically defined tunneling barrier. The top portion of each frame in Fig. 3 depicts the one-dimensional channel along which the electrons travel and interact. At the center, the channel width is narrower, giving rise to a tunneling barrier. The barrier can arise from both quantum confinement along the transverse direction and from the electrostatically defined linewidth. The bottom portion shows the electrostatic potential along the center of the nanowire. Each of the two potential wells holds a single electron. Also shown are the two lowest-energy solutions to the one-dimensional Schrödinger equation. Within the single-electron approximation, these solutions indicate the probability distribution for the electrons and their response to the transient coupling potential. The electrons are brought together using transient fields from a pair of femtosecond pulses centered on either side of the tunneling barrier. As these fields decay, a weaker third pulse is applied to the center of the barrier region, ensuring that the electrons return to their bound states.

The exchange operator $\hat{U}_{ex}(\theta)=\exp[-i\theta\hat{\mathbf{S}}_1\cdot\hat{\mathbf{S}}_2]$ is characterized by an angle θ (see Ref. [9]) such that $\theta=\pi$ corresponds to a ''swap'' operation. In order to implement exchange-only universal gates, it is necessary to vary θ continuously from 0 to 2π. Such control can be achieved by varying both the strength and duration of the optical pulses. Long-range coupling is provided through the use of sequentially applied swap operations.

In constructing an exchange gate such as the one in Fig. 3, it is important to consider possible decoherence mechanisms [35]. For instance, if the adiabatic condition is relaxed, the loss of spatial coherence can lead to decoherence of the spin wave function. One method of reducing such effects is to couple the spins via a superexchange mechanism [36]. By forcing electrons to communicate through a single-quantum channel, such effects can be greatly reduced.

B. Quantum-information processing

Once the basic one-qubit and two-qubit operations are established, it will be important to ''program'' the quantum-information processor to perform complex combinations of such operations. In recent years, Keith Nelson and coworkers at MIT have developed the optical technology for converting a femtosecond seed pulse into a controlled pattern of pulses shaped in both space and time [31]. At the core of this technology is a commercially available two-dimensional spatial light modulator (Hamamatsu PAL-SLM) that can be programmed using the standard visual graphic adapter output of a PC video card. The algorithms for producing controlled waveforms of a desired pattern have also been developed [37]. At present, the speed with which the array could be reprogrammed limits the interactive use of the SLM in a quantum calculation (the speed is quite slow, ~10 ms); however, if a repetitive calculation is performed, then it will not be necessary to reprogram continually the SLM.

V. SUMMARY

I have described a method for controlling electron spin interactions in silicon-based structures using the static and dynamic polarization of an epitaxial ferroelectric thin film. Solid state approaches to quantum-information processing and quantum computing have many advantages. In particular, silicon-based approaches are especially attractive, given the natural interface to classical computing architectures. The question of whether it will be possible to satisfy the physical requirements for quantum computation can only be answered through experimental investigation.

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