Understanding Charge Dynamics in Silicon Dangling Bond Structures for Nanoscale Devices

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ABSTRACT

A dangling bond (DB) on a silicon surface atomic has been found to behave as an atomic-scale quantum dot. This opens up the possibility of using DBs as building blocks for novel electronic structures, pushing the ultimate limits of nanoscale devices. Examples include: artificial molecules, nanowires, and alternate computing architectures such as the Quantum-Dot Cellular Automata (QCA). We study theoretically various aspects of DB structures on an H-terminated 2x1 Si(001) surface, which are of primary interest for the advancement of any device architecture: (i) The "diatomic" artificial DB molecule, composed of two tunnel-coupled DBs; its coherence and polarization properties in different ionic states; its potential as a charge qubit. (ii) The interaction of such DB structures with external control electrodes and manipulators, including the interaction with a scanning tunneling microscopy (STM) probe.

Keywords: coherent electron dynamics, computing architectures, charge manipulation and control.

1 MOTIVATION

As the need of miniaturization and low power consumption in today's electronic devices continuously increases so does the interest in structures known as artificial atoms and molecules. These are fabricated nanostructures mimicking "natural" atoms and molecules, which confine electrons in all three spatial directions, attempting to exploit the electronic properties and functions of matter at the nanoscale. Perhaps the most important physical realizations of these concepts are found in structures composed of metallic and semiconductor quantum dots (QD). Thus, single QDs can be seen as artificial atoms, while a set of coupled QDs can behave as an artificial molecule. Recently, silicon surface dangling bonds (DBs) have been found to behave as atomic-scale quantum dots in that they hold a discrete number of localized electrons with well defined eigen-energy and spatial eigen-function. Most remarkable about using silicon DBs as QD is the extreme down-scaling of electron localization, virtually approaching the true atomic scale. Furthermore, their fabrication and manipulation is facilitated by today's scanning probe microscopy apparatus. As complex structures of such DBs become available on silicon surfaces, their characterization and understanding - in a device-functional sense - emerges as an imperative theoretical task. Due to the nature of such systems, electron correlation plays an important role in determining device properties, and the use of many-body physics methods is a necessity.

2 DANGLING BONDS AS ELECTRONIC EIGEN-STATES

A silicon surface DB is an electronic state confined to the vicinity of a host Si atom with an unsatisfied bond, and surrounded on the surface by other hydrogen-terminated Si atoms. A DB can host zero, one, or two electrons, depending on the filling conditions given by its environment, more precisely, by the Fermi level of the host crystal. In turn, the DB eigen-energy and wavefunction depend on its filling, yielding three distinct energy levels $E(DB^{+})$, $E(DB^{0})$, and $E(DB^{-})$, respectively for each filling state.

Calculating the exact form of the eigen-function for the DB is generally a complex task and involves taking into account all the interactions with the crystal electrons and ions. As a consequence, there is a dependence of the eigen-energy on the particular type of surface reconstruction of the host silicon crystal. To date, there are only approximate theoretical results for the DB eigen-energy, roughly corroborated by experiments. In this study we rely on previously published DFT calculations from our group [1-3].

2.1 The form of DB orbitals

In this study, we assume a simple semi-empirical form of the DB orbital, namely a modified Slater-type orbital (MSTO). The main modification is introduced to account for the anisotropy of the ionization potential due to the presence of the surface [4].

$$\psi_{DB}(r,\theta,\varphi) = Cr^{n-1}\exp[-\zeta(\theta)r]Y_l^m(\theta,\varphi), \qquad (1)$$

where the decay rate $\zeta(\theta)$ depends on the polar angle

$$\zeta(\theta) = \sqrt{2mW(\theta)}/\hbar \ , \qquad (2)$$

and where W is the ionization potential *function* given in a phenomenological form accounting for the transition between vacuum and silicon bulk:

$$W(\theta) = W_b + (W_v - W_b)[\tanh(\zeta_0 r\cos\theta) + 1]/2\,, \quad (3)$$

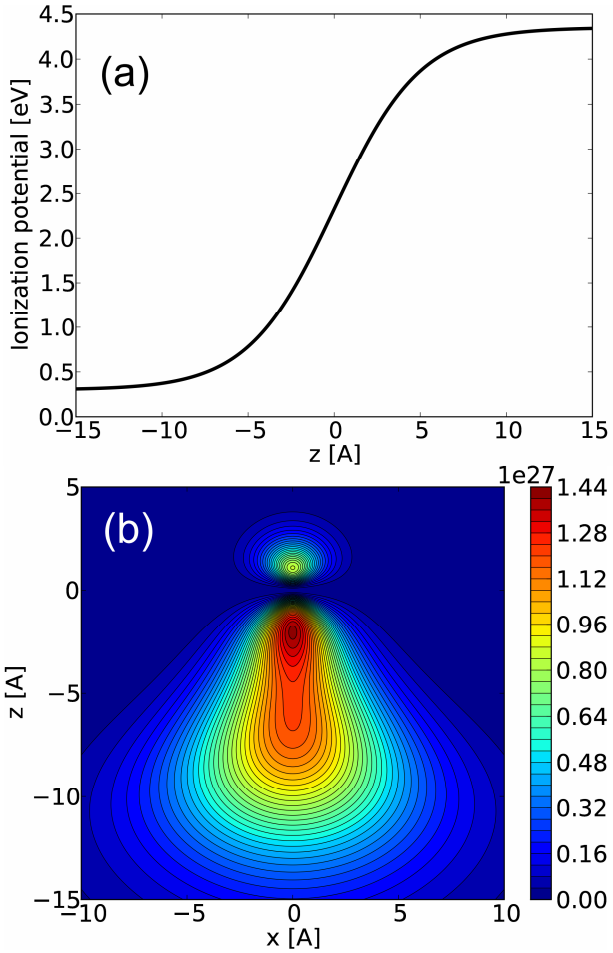

Figure 1: The DB$^-$ orbital: (a) the ionization potential as a function of surface-perpendicular distance (positive z in the vacuum). (b) Charge density of a DB in the axial plane as predicted by the modified STO in eq. (1). The color bar indicates the values of the density in units of 10^{27} m^{-3}.

where $W_{b,v}$ are the ionization potentials toward bulk and vacuum respectively, and ζ_0 is a decay constant. The results of such a choice for the wavefunction are depicted in Figure 1.

2.2 Tunnel-Coupled DB pairs

Closely positioned DBs can become tunnel-coupled and thus form new molecular-type orbitals. Such coupled DB structures are thus analogous to artificial molecules. On a typical H-Si (001) 2x1 surface, coupling occurs when DBs are within ~ 2nm of each other and when the Coulomb interaction precludes full (double) occupation of the DBs involved.

Electron correlation can play an important role in DB structures, therefore many-body models are useful in their description. The simplest and most prominent model in this case is the Hubbard model and its variations. We employ here an extended Hubbard model which incorporates electron-electron interactions, as well as external perturbations on the system. The model Hamiltonian reads

$$H = \sum_{i,\sigma}(E_0 - \mu)n_{i,\sigma} + \sum_{i,\sigma}V_{ext}n_{i,\sigma} + \sum_{i}Un_{i,\sigma}n_{i,-\sigma} - \sum_{i<j,\sigma}t_{i,j}(c^+_{i,\sigma}c_{j,\sigma} + c^+_{j,\sigma}c_{i,\sigma}) + \sum_{i<j,\sigma,\tau}V_{e-e}n_{i,\sigma}n_{j,\tau} \quad (4)$$

where E_0 is the self-energy, μ the chemical potential, t_{ij} are the hopping integrals, U the Hubbard on-site repulsion, V are interaction energy terms; n, c^+, c are the occupation, creation, and anihilation operators, respectively. Select values for the model parameters are given in Table 1. The interaction of a DB structure with an external electrode (e.g. a biased STM tip) is incorporated in our model via the second term in eq. (4).

DB separation[Å]	t[meV]	$V_{e\text{-}e}$[meV]
3.86	307.7	0.3732
7.72	87.8	0.2358
11.58	43.8	0.1650
15.44	18.9	0.1262
19.30	4.6	0.1020
11.77	42.3	0.1626
9.82	62.0	0.1915

Table 1: Model parameters for the extended Hubbard Hamiltonian used to describe coupled DBs. The discrete DB separation is imposed by the surface lattice geometry. t is the hopping integral and $V_{e\text{-}e}$ is the Coulomb interaction between DBs.

We also use a different model for our system, namely the molecular orbital approach in quantum chemistry, in order to compare with the above model and thus assess the role of electron correlations in DB structures.

3 QCA DEVICES

Quantum-Dot Cellular Automata [5] is an alternate information processing paradigm, that employs the electronic orbital degree of freedom, without actually carrying current through the device. In a QCA device, information processing is mediated via the Coulomb interaction between neighboring electrons.

DB structures are ideal for implementing QCA devices because their extreme miniaturization makes the Coulomb interaction much greater than the thermal energy and thus make information processing robust at room temperature. The challenge resides in addressing the individual electrons with appropriately sized contacts and suitable control apparatus. The behaviour of a QCA device implemented

with silicon DBs can be captured by the extended Hubbard model outlined above. Some of the model parameters are extracted from first principles quantum mechanical calculations, such as the self-energies and hopping integrals. Another important ingredient of the model is the interaction between the DB electrons and the substrate, in particular, the effects of screening due to mobile charge carriers in the semiconductor. To this end, finite element methods are used to solve for the electrostatic potential in a self-consistent way.

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