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Proposal for electron spin relaxation measurements using double-donor excited states in Si quantum computer architectures

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The possibility of performing single-spin measurements in Si-based quantum computers using electrons bound to double donors near a barrier interface is assessed. We find that double donors in the ground state require electric fields and tunneling times too large for practical implementations, while operations with double donors in their first excited state involve smaller fields and faster tunneling times, and are therefore suitable for spin-to-charge conversion measurements. We propose a measurement scheme that would render ensemble estimates of the spin coherence at the Si/SiO$_2$ interface.

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Among the several operations required for a spin-based quantum computer (QC), single-spin rotation and measurement are probably the hardest ones to achieve. Not only is the electron spin extremely weak and, therefore, hard to detect,[1] operation and measurement times must be fast enough compared to electronic spin dephasing times. Single-electron-spin control has very recently been demonstrated in a double-quantum-dot configuration in a GaAs heterostructure by Koppens *et al.*[2] In the occupation number representation for the individual dots, the involved states are (1,1), one electron in each dot, and (0,2), one of the dots empty and the other doubly occupied. Due to the Pauli principle, the state (0,2) is accessible only if the electrons form a spin singlet, which allows inferring spin states from charge transport measurements.[2] Although a similar experiment has been suggested using double donors in Si,[3] and other approaches have been attempted[4] for single-donor-based QC in Si,[5] progress in donor-based Si systems has been much slower than in the GaAs quantum dot systems, primarily because practical schemes for spin-to-charge conversion have not been theoretically proposed in these architectures. The proposal by Kane *et al.*[3] has many of the basic ingredients that led to the recent successful results in GaAs quantum dots: The double-dot potential would correspond to a double-well structure formed by a double donor (DD) in Si near the interface with a barrier (e.g., Si/SiO$_2$) under a uniform electric field applied perpendicular to the interface. The (1,1) state would correspond to one electron in the characteristic triangular-shaped interface-plus-electric-field well and one in the double-donor Coulomb well, while the (0,2) state would correspond to both electrons bound to the (neutral) DD.

We explore here the use of DDs in Si (solid-state analogs of the He atom) to perform one-qubit operations similar to those reported in Koppens *et al.*'s experiments. For the measurement of a single spin state, we quantitatively estimate the required electric fields for the first ionization of Te in Si, assessing the practical implementation of the scheme in Ref. 3. We also present a related proposal for the measurement of spin relaxation times in Si near a SiO$_2$ interface, a key parameter if qubit measurement is to be performed at the interface.[5] The proposal here is similar to the one that has allowed the measurement of spin dephasing times in double quantum dots with two electrons in GaAs.[6] In this case we show that S double donors would be the most appropriate due to the negligible spin-orbit coupling.

Double donors (S, Se, Te) in Si are substitutional deep centers whose electrons' binding energies (summarized in Table I) are typically one order of magnitude larger than for single donors (P, As, Sb). S, Se, and Te are all slow diffusers in Si (Ref. 7) and, therefore, appropriate for donor-based Si QC processing techniques. The substitutional double-donor ground state is a spin singlet analogous to the He atom ground state.[8] The particular band structure of Si and the local tetrahedral symmetry of the potential leads to an orbital ground state for substitutional donors of A_1 symmetry. The first excited state for DDs is such that one of the electrons is in the ground state and the other in the next-in-energy $1s(T_2)$ orbital state.[9] This outer electron experiences a closely hydrogenic effective potential because the DD core charge is virtually screened by the inner $1s(A_1)$ electron,[9] which has a very small effective Bohr radius. The corresponding spin states are always singlet for the ground state, while the first excited configuration can be in a triplet or a singlet, with the triplet lower in energy due to Hund's rule. The binding energies of the ground and the first excited states of S, Se, and Te are shown in Fig. 1.

We consider a DD on its ground state in Si, a distance d away from a (001) interface with SiO$_2$. When a characteristic electric field F_c along z (perpendicular to the interface) is applied, one of the electrons may tunnel toward the interface well [where it still experiences the Coulomb attraction of the donor, keeping it from spreading in a two-dimensional elec-

TABLE I. Energy of neutral ($E_{\rm DD}$) and singly ionized ($E_{\rm DD^+}$) double donors. The first ionization energy is $E_{\rm DD^+}-E_{\rm DD}$ (Ref. 11).

Donor	$E_{\rm DD}$ (meV)	$E_{\rm DD^+}$ (meV)	First ionization (meV)
Te	609.6	410.8	198.8
Se	899.9	593.3	306.6
S	931.8	613.5	318.3

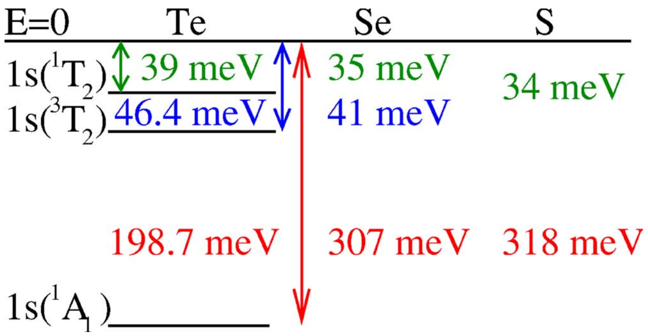

FIG. 1. (Color online) First ionization energies of the $1s$ levels of Te, Se, and S in Si (Refs. 11 and 12). The ground state is the singlet $1s(^1A_1)=[1s(A_1),1s(A_1)]$ and the first excited states are $1s(^iT_2)=[1s(A_1),1s(T_2)]$. $i=1$ (3) corresponds to a singlet (triplet).

tron gas (2DEG)].[10] Small changes of the field around F_c cause the electron to move between the donor and the interface. This "shuttling" is allowed only as long as the two electrons are in a spin-singlet state, while if they are in a spin-triplet state the electron cannot tunnel back to the donor due to the Pauli principle. The electron motion can be detected by single-electron transistors on the device surface, above the oxide layer.[3] As a result, the measurement of charge leads to information about the spin state of the electron (spin-to-charge conversion).

As shown in Table I, Te is the shallowest of the DDs in Si, with a total (two-electron) binding energy $E_{\rm Te}=609.6$ meV. For this reason it is the most appropriate for electric-field-driven ionization experiments in Si, as it requires the smallest field $F_c^{\rm Te}$ to ionize. $F_c^{\rm Te}$ has been estimated, within a single-electron approach, in Ref. 3. Here we consider the full two-particle Hamiltonian, including the electron-electron interaction and the image charges, to calculate $F_c^{\rm Te}$ as a function of d.

We solve the two-electron double-donor problem within several simplifying approximations, namely, the single-valley effective mass approach, assuming an isotropic effective mass m^*. The two-particle Hamiltonian in atomic units ($a^*=\hbar^2\epsilon_{\rm Si}/m^*e^2$ and ${\rm Ry}^*=m^*e^4/2\hbar^2\epsilon_{\rm Si}^2$) for a DD in Si close to a Si/SiO$_2$ (0,0,1) interface in the presence of an electric field F is

$$H=T_1+T_2-\frac{4}{r_1}-\frac{4}{r_2}+\kappa eF(z_1+z_2)+\frac{2}{r_{12}}+V_{\rm image}^D+V_{\rm image}^e, \tag{1}$$

with kinetic energy $T_i=-\partial^2/\partial x_i^2-\partial^2/\partial y_i^2-\partial^2/\partial z_i^2$ $(i=1,2)$, $\kappa=3.89\times10^{-7}\epsilon_{\rm Si}^3(m/m^*)^2$ cm/kV, and F in kV/cm. We estimate the isotropic radius $a_{\rm Te}$ and mass m^* from the experimental value of the ground-state energy of Te, $E_{\rm Te}=-609.6$ meV, using the calculated expression for He, $E_{\rm Te}=2/a_{\rm Te}^2-8/a_{\rm Te}+5/4a_{\rm Te}=-5.6953$ Ry* minimized by $a_{\rm Te}=0.59a^*$.[8] The gives ${\rm Ry}^*=107$ meV, $m^*\sim m$, and $a^*=0.6$ nm. The image terms are

$$V_{\rm image}^D=\frac{4Q}{\sqrt{x_1^2+y_1^2+(z_1+2d)^2}}+\frac{4Q}{\sqrt{x_2^2+y_2^2+(z_2+2d)^2}}, \tag{2}$$

the interaction of the electrons with the DD image, and

$$V_{\rm image}^e=-\frac{Q}{2(z_1+d)}-\frac{Q}{2(z_2+d)}-\frac{4Q}{\sqrt{(x_1-x_2)^2+(y_1-y_2)^2+(z_1+z_2+2d)^2}}, \tag{3}$$

the interaction of the electrons with their own and the other electron's images. $Q=(\epsilon_{\rm SiO_2}-\epsilon_{\rm Si})/(\epsilon_{\rm SiO_2}+\epsilon_{\rm Si})$, with $\epsilon_{\rm Si}=11.4$ and $\epsilon_{\rm SiO_2}=3.8$.

We solve the Hamiltonian in the nonorthogonal basis defined by the initial state $\Psi_A\equiv(0,2)$ with the two electrons in the donor ground state $[1s(A_1),1s(A_1)]$: $\Psi_A=\psi_D(1)\psi_D(2)$, and the final singly ionized donor state and one electron at the interface $\Psi_B\equiv(1,1)$: $\Psi_B=[\psi_D(1)\psi_I(2)+\psi_D(2)\psi_I(1)]/\sqrt{2(1+S_{ID}^2)}$ where $\psi_D(i)\propto e^{r_i/a_{\rm Te}}$, $\psi_I(i)\propto(z_i+d)^2e^{-\alpha(z_i+d)/2}e^{-\beta^2\rho_i^2/2}$,[13] $i=1,2$, and $S_{ID}=\langle\psi_D|\psi_I\rangle$ is the overlap between the single-electron states. The variational parameters $a_{\rm Te}$, α, and β minimize the expectation value of the energy and lead to approximate expressions for the one-electron ground states ψ_D and ψ_I at the donor well and the interface well, respectively.

The Bohr radius of the DD ground state ($a_{\rm Te}=0.354$ nm) is one order of magnitude smaller than the one for P ($a_{\rm P}=2.365$ nm) due to the larger binding energy allowing two useful simplifications: (i) the overlap S_{ID} is very small, therefore the exchange part of the electron-electron interaction can be neglected; (ii) in the limit $d\gg a_{\rm Te}$ the electron at the interface sees a nuclear image with charge +1 (H-like atom) rather than a +2 donor plus an electron (first ionized He-like atom). Hence, all the image terms [Eqs. (2) and (3)] in the Hamiltonian are reduced to

$$V_{\rm image}^e+V_{\rm image}^D\approx-\frac{Q}{2(z_1+d)}-\frac{Q}{2(z_2+d)}+\frac{2Q}{\sqrt{x_1^2+y_1^2+(z_1+2d)^2}}+\frac{2Q}{\sqrt{x_2^2+y_2^2+(z_2+2d)^2}}, \tag{4}$$

and the variational parameters for $\psi_I(i)$ are the same as for the single-donor problem.[10]

We calculate $F_c^{\rm Te}$ [shown in Fig. 2(a)] from the condition $\langle\Psi_A|H|\Psi_A\rangle=\langle\Psi_B|H|\Psi_B\rangle$. The results are similar to the single-electron approach:[3] when the DD is located at $d=20$ nm, $F_c^{\rm Te}=190$ kV/cm. These large values would probably cause electrical breakdown in the nanostructures. The lower curves in Fig. 2(a) correspond to the single-donor P results[10] and are shown for comparison. The curves correspond to isotropic and anisotropic Ψ_D wave functions, respectively, and are very similar, validating the isotropic approximation adopted here for the DD problem. In Fig. 2(b) we reproduce the tunneling times $\tau_{\rm P}$ required to ionize P.[10] τ is inversely proportional to the overlap between states A and B, which is orders of magnitude smaller in the DD problem than in the single-donor problem. Therefore, we expect $\tau_{\rm Te}$ to be orders of magnitude larger than $\tau_{\rm P}$ and hence probably of the same order as the spin decoherence time in bulk natural

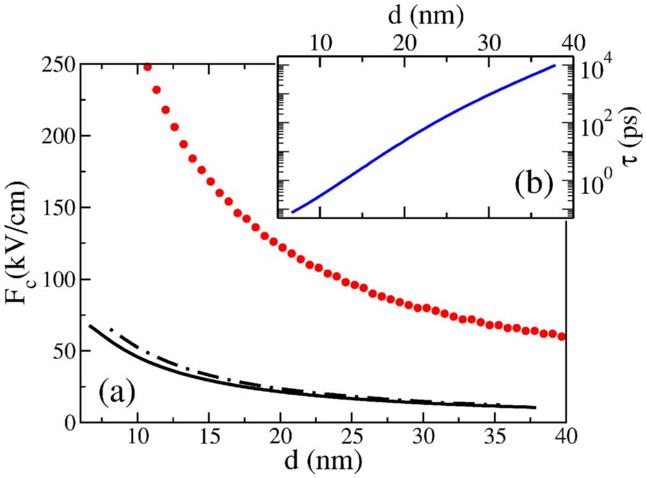

FIG. 2. (Color online) (a) Electric field F_c^{Te} (circles) required to take one donor electron to the interface. We compare it to the single-donor (P) results with an anisotropic donor wave function (Ref. 10) (solid line), and with an isotropic donor wave function (dot-dashed line) with $m^*=0.298m$. (b) Tunneling time required to ionize an electron in the first excited state of a double donor (the same as required to ionize the single donor P) (Ref. 10).

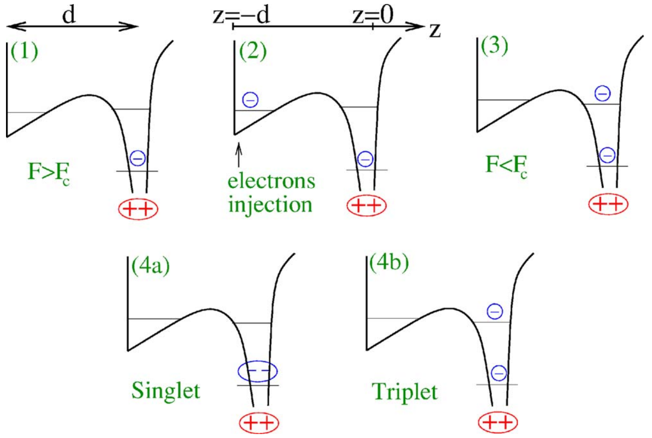

FIG. 3. (Color online) Double donors in their first excited state can be used to measure the spin dephasing times in Si close to an interface. See text for details.

Si (∼1 ms). Consequently, DDs in the ground state are of no practical use to single-spin measurements.

This is not the case if the DD is in its first excited $[1s(A_1),1s(T_2)]$ state, which would be more directly accessible for electric-field-driven $(1,1)\leftrightarrow(0,2)$ manipulations. In contrast to the large binding energy and small Bohr radius of the deep-center ground state, the outer electron in the first excited states of neutral DDs has similar properties[9] (binding energy ∼32 meV and Bohr radius ∼2 nm) as an electron in a single-donor ground state.[14] Hence, F_c required to singly ionize $[1s(A_1),1s(T_2)]$ and the tunneling times involved in this process are similar to the ones calculated for P, in Fig. 2, and, therefore, experimentally meaningful.[10] We propose a scheme to measure the spin dephasing times T_2^* involving DDs in their first excited state. As opposed to previous works, which have measured spin dephasing times of electrons at donors in bulk Si,[15] at donors 50 nm from the interface,[16] and in a 2DEG in Si/SiGe structures,[17] our proposed experiment would measure the spin dephasing time of confined electrons at the Si/SiO$_2$ interface.

Figure 1 shows the first ionization energy for the ground state (singlet) and the first excited state (singlet or triplet) of the neutral DDs. The first excited states have been observed in absorption spectra from the ground state $[1s(A_1),1s(A_1)]$.[12,18] Although transitions to pure spin-triplet states are forbidden by selection rules, they have been observed in Te and Se:[12,18] The spin is not a good quantum number due to spin-orbit interaction. On the other hand, only the spin-singlet state has been observed in S, due to the much smaller spin-orbit interaction for the lighter donor. The spin-triplet-state lifetimes in Te and Se are of the order of tens of picoseconds as estimated from the linewidths (which could be limited by experimental precision),[18] not much longer than the spin-singlet-state lifetimes. The spin-singlet lifetime in S is expected to be of the same order as for Te and Se, while the spin-triplet lifetime must be much longer due to the relatively small spin-orbit interaction.[19] Moreover, it has been recently reported[20] that the linewidths of absorption transitions in P-doped Si can be decreased (and, hence, the lifetimes increased) by using isotopically purified Si, and it is expected that the linewidths of deeper impurities (like S) could be even more dramatically reduced due to the smaller Bohr radius.[21] Therefore, in S the first excited triplet state lifetime is expected to be much longer than the first excited singlet state lifetime. We consider now Si doped with S.

For S-doped Si, it has been observed experimentally that about 50% of the donors remain singly ionized[11] [their only electron is in the inner $1s(A_1)$ state] while the others remain neutral and inert for this proposal. We could use such singly ionized donors to measure the spin dephasing time by manipulating their outer electrons between the interface and the sulfur $1s(T_2)$ orbital (under a magnetic field <1 T to define a quantization axis) in the way illustrated in Fig. 3. (1) An electric field slightly larger than the F_c required to ionize a donor in its first excited state is applied. (2) Electrons are injected at the interface (for instance, by using heavily doped contacts). The ionized donors bind the new electrons at the interface[10] leading to a (1,1) configuration. (3) The electric field is decreased so that, at F_c, the electron at the interface goes to the outer $1s(T_2)$ state at the donor [configuration (0,2)] with typical tunneling times as in Fig. 2(b). (4a) If the two-electron spin state is a singlet, or contains a significant singlet component, the orbital state will rapidly decay to the ground state $[1s(A_1),1s(A_1)]$; (4b) while if they form a triplet, selection rules imply a much longer-lived state. The electric field is increased again, taking the outer electron at the donor back to the interface. Only those electrons whose spins form a triplet with the inner electron will remain in the excited state long enough to be able to shuttle $[(1,1)\leftrightarrow(0,2)]$ when the electric field dithers around F_c. The inner electron remains bound to the donor because the field required to doubly ionize the donor is much larger ($\gg F_c^{\mathrm{Te}}$). Detection of charge at the interface will give a decreasing population of electrons, from which we can extract the spin dephasing time of electrons at the Si/SiO$_2$ interface.

There are various time scales involved in this problem: the unknown spin-triplet lifetime of the first excited state in S (which we have argued should be orders of magnitude longer than the spin-singlet ~ picosecond lifetime), the tunneling times for the electron shuttling [as given in Fig. 2(b)], and the frequency of the electric field dithering (with corresponding times $\gtrsim 1$ ns). Assuming the spin-triplet lifetime is longer than the other two time scales, the proposed experiment would measure a combination of spin-triplet lifetime and spin dephasing T_2^*, giving a lower bound to T_2^*. These measurements would also give estimates for the donor first excited state spin-triplet lifetime. It is reasonable to assume that the spin coherence time at interfaces can be made much longer than any of the other time scales by improving the interface quality (by using, for instance,[16] hydrogen-passivated Si instead of SiO_2) and by isotopically purifying the Si host. Note that the already long bulk value ~1 ms can be dramatically increased rather easily to 100 ms or longer by isotopic purification.[15,22]

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[1] D. Rugar, R. Budakian, H. J. Mamin, and B. W. Chui, Nature (London) **430**, 329 (2004); M. Xiao, I. Martin, E. Yablonovitch, and H. Jiang, *ibid.* **430**, 435 (2004).

[2] F. H. L. Koppens, C. Buizert, K. J. Tielrooij, I. T. Vink, K. C. Nowack, T. Meunier, L. P. Kouwenhoven, and L. M. K. Vandersypen, Nature (London) **442**, 766 (2006).

[3] B. E. Kane, N. S. McAlpine, A. S. Dzurak, R. G. Clark, G. J. Milburn, H. B. Sun, and H. Wiseman, Phys. Rev. B **61**, 2961 (2000).

[4] K. R. Brown, L. Sun, and B. E. Kane, Appl. Phys. Lett. **88**, 213118 (2006); A. R. Stegner, C. Boehme, H. Huebl, M. Stutzmann, K. Lips, and M. S. Brandt, Nat. Phys. **2**, 835 (2006); H. Sellier, G. P. Lansbergen, J. Caro, N. Collaert, I. Ferain, M. Jurczak, S. Biesemans, and S. Rogge, Phys. Rev. Lett. **97**, 206805 (2006); L. C. L. Hollenberg, A. S. Dzurak, C. Wellard, A. R. Hamilton, D. J. Reilly, G. J. Milburn, and R. G. Clark, Phys. Rev. B **69**, 113301 (2004); L. C. L. Hollenberg, C. J. Wellard, C. I. Pakes, and A. G. Fowler, *ibid.* **69**, 233301 (2004).

[5] B. E. Kane, Nature (London) **393**, 133 (1998).

[6] J. R. Petta, A. C. Johnson, J. M. Taylor, E. A. Laird, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Science **309**, 2180 (2005); F. H. L. Koppens, J. A. Folk, J. M. Elzerman, R. Hanson, L. H. W. van Beveren, I. T. Vink, H. P. Tranitz, W. Wegscheider, L. P. Kouwenhoven, and L. M. K. Vandersypen, *ibid.* **309**, 1346 (2005); A. C. Johnson, J. R. Petta, J. M. Taylor, A. Yacoby, M. D. Lukin, C. M. Marcus, M. P. Hanson, and A. C. Gossard, Nature (London) **435**, 925 (2005).

[7] *Semiconductors—Basic Data*, 2nd ed., edited by O. Madelung (Springer, Berlin, 1996).

[8] J. C. Slater, *Quantum Theory of Molecules and Solids* (McGraw-Hill, New York, 1963), Vol. 1.

[9] T. Ning and C. Sah, Phys. Rev. B **4**, 3482 (1971).

[10] M. J. Calderón, B. Koiller, X. Hu, and S. Das Sarma, Phys. Rev. Lett. **96**, 096802 (2006).

[11] H. Grimmeis and E. Janzén, in *Deep Centers in Semiconductors*, edited by S. Pantelides (Gordon and Breach, New York, 1992), Chap. 2, p. 87.

[12] K. Bergman, G. Grossmann, H. G. Grimmeiss, M. Stavola, C. Holm, and P. Wagner, Phys. Rev. B **37**, 10738 (1988).

[13] M. J. Calderón, B. Koiller, and S. Das Sarma, Phys. Rev. B **75**, 125311 (2006).

[14] W. Kohn, *Shallow impurity states in silicon and germanium*, in Solid State Physics Series, Vol. 5, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1957), pp. 257–320.

[15] A. M. Tyryshkin, S. A. Lyon, A. V. Astashkin, and A. M. Raitsimring, Phys. Rev. B **68**, 193207 (2003); E. Abe, K. M. Itoh, J. Isoya, and S. Yamasaki, *ibid.* **70**, 033204 (2004); A. M. Tyryshkin, J. J. L. Morton, S. C. Benjamin, A. Ardavan, G. A. D. Briggs, J. W. Ager, and S. A. Lyon, J. Phys.: Condens. Matter **18**, S783 (2006).

[16] T. Schenkel, A. M. Tyryshkin, R. de Sousa, K. B. Whaley, J. Bokor, J. A. Liddle, A. Persaud, J. Shangkuan, I. Chakarov, and S. A. Lyon, Appl. Phys. Lett. **88**, 112101 (2006).

[17] A. M. Tyryshkin, S. A. Lyon, W. Jantsch, and F. Schaffler, Phys. Rev. Lett. **94**, 126802 (2005).

[18] R. E. Peale, K. Muro, A. J. Sievers, and F. S. Ham, Phys. Rev. B **37**, 10829 (1988).

[19] The atomic values of the one-electron spin orbit parameters are 558 (Te), 278 (Se), 64 (S), and 24 meV (Si) [F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall, Englewood Cliffs, NJ, 1963)]. For substitutional donors the values are 1.5375 (Te) and 0.3967 meV (Se) (the value for S was too small to be detected) (see Ref. 12).

[20] D. Karaiskaj, J. A. H. Stotz, T. Meyer, M. L. W. Thewalt, and M. Cardona, Phys. Rev. Lett. **90**, 186402 (2003).

[21] M. Cardona and M. Thewalt, Rev. Mod. Phys. **77**, 1173 (2005).

[22] W. M. Witzel, R. de Sousa, and S. Das Sarma, Phys. Rev. B **72**, 161306(R) (2005).