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Permittivity of oxidized ultra-thin silicon films from atomistic simulations

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Abstract—We establish the dependence of the permittivity of oxidized ultra-thin silicon films on the film thickness by means of atomistic simulations within the density-functional-based tight-binding theory (DFTB). This is of utmost importance for modeling ultra- and extremely-thin silicon-on-insulator MOSFETs, and for evaluating their scaling potential. We demonstrate that electronic contribution to the dielectric response naturally emerges from the DFTB Hamiltonian when coupled to Poisson equation solved in vacuum, without phenomenological parameters, and obtain good agreement with available experimental data. Comparison to calculations of H-passivated Si films reveals much weaker dependence of permittivity on film thickness for the SiO₂-passivated Si, with less than 18% reduction in the case of 0.9 nm silicon-on-insulator.

Index Terms—permittivity, atomistic modeling, oxide interface, density-functional tight binding, silicon-on-insulator

It is well known that the dramatic reduction of the dimensions of the Si channel, e.g. in ultra-thin-body silicon-on-insulators devices, leads to a significant change in the electronic and dielectric properties of Si, particularly at channel thickness below 6 nm [1–3]. A number density-functional theory (DFT) studies applied to hydrogen-passivated Si films suggest that the decrease in permittivity with the decrease of Si-film thickness becomes significant even earlier than the corresponding widening of the fundamental band-gap, and predict 35–45% reduction at around 1 nm [4–6]. On the experimental side however, we are aware of only one study of oxidized Si films down to 3.3 nm, and while a qualitative trend is evident, the scatter of the results precludes us from establishing an accurate quantitative picture towards sub-nm Si thickness [7]. We note further that to the best of our knowledge, the permittivity dependence of oxidized Si films has not been modeled *ab initio*, most likely due to the complexity and cost associated with including the oxide on each side of Si in DFT. However, very recently we

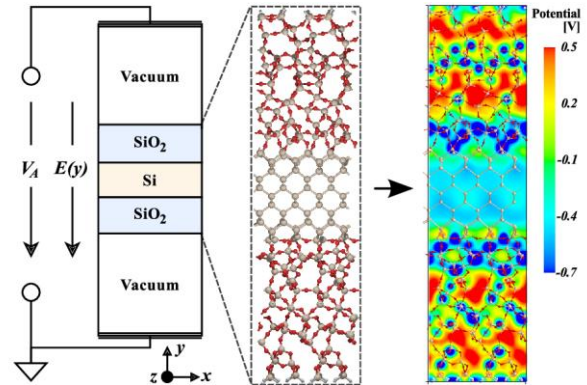


Fig. 1. Schematic of the simulation setup; atomic structure of a SiO₂-Si-SiO₂ periodic model with 1.4 nm Si, repeated 2 times in x and z ; and the corresponding color-map of the potential in a plane orthogonal to the z axis, showing large fluctuations around the atoms at the interface and in the oxide.

demonstrated that density-functional tight-binding theory (DFTB), with an accurate parameterization, can give us very good description of the electronic properties not only of bulk Si and SiO₂, but also of their interface [3], and allows us to explore substantially larger systems, including transport through ETSOI devices [8]. The purpose of this paper is therefore twofold: 1) to evaluate a way of calculating the dielectric constant of thin-films within the framework of DFTB; and 2) to establish the permittivity dependence on the thickness of oxidized ultra-thin Si films.

We employ the DFTB+ computer code [9], implementing the self-consistent-charge DFTB, coupled self-consistently to a Poisson solver [10–12]. This permits us to apply bias (V_A) and find the distribution of potential (ϕ) and electric field (E) in the model atomic structures. The atomic models are the SiO₂/Si/SiO₂/vacuum super-cells with varying Si thickness and 2.2 nm amorphous SiO₂ used in our recent study [3]. Ground-state calculations are performed at 0 K, with $8 \times 1 \times 8$ Monkhorst-Pack sampling grid of the Brillouin zone. The approach for evaluating the dielectric constant is conceptually summarized in Fig. 1(a) and has been used in other studies [13][5]. It is based on the continuity of electric displacement vector, which for a linear and isotropic model system at 0 K, without free carriers, translates to $\epsilon_i E_i(y) = \epsilon_{i+1} E_{i+1}(y)$, where i and $i+1$ label two neighboring layers, ϵ_i and E_i being the permittivity and electric field. Considering the five-layer system of Fig. 1 and knowing the vacuum permittivity, ϵ_0 , we can find the *macroscopic* permittivity of SiO₂ and Si, if we determine the *macroscopic* field across each layer. This is not

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straightforward, considering the rapid fluctuations of the potential, as also shown in Fig. 1. These fluctuations average out in the planes parallel to the interfaces but persist along the interface-normal (y -direction) as seen in Fig. 2 for two different values of applied bias. It should be understood that the potential from DFTB reflects only the net atomic charges projected on the Poisson grid via exponentials with atom-specific rate that corresponds to the Hubbard value of the chemical element [10][12]. These are charge fluctuations that arise from charge transfer, i.e. bond-asymmetry around the interface and in the oxide, and induced polarization, i.e. external electric field. The charge transfer component is dominant by far and is common regardless of applied bias (structure is not relaxed under bias), and is shown in Fig. 2. Subtracting it from the total fluctuations at non-zero bias yields the induced charge, shown in Fig. 3(a). If we similarly take the difference between the potential profiles of Fig. 2, i.e. with and without applied bias, we obtain the potential difference $\Delta\phi$, also shown in Fig. 3(a). We see that $\Delta\phi$ is readily interpreted as a macroscopic potential as it decays apparently linearly – with different rate in each of the five layers of our system. Therefore, the negative gradient of $\Delta\phi$ yields the electric field, which is shown in Fig. 3(b). It naturally shows smooth transition from one layer to another, suggesting gradual change in permittivity from Si to SiO₂. The layer-averaged, *macroscopic* fields are also shown in Fig. 3(b), and for the given case of 1.4 nm Si and 2.2 nm SiO₂ we evaluate dielectric constants of 9.8 and 2.7.

Applying the continuity of the displacement field on a *microscopic* level we obtain the permittivity profile $\varepsilon(y)$, shown in Fig. 4(a), (thin line). Smoothing with a Gaussian kernel with a standard deviation of 2.3 Å almost eliminates the atomic fluctuations as shown (thick line). Averaging the *inverse* permittivity in each material layer yields the value obtained from the *macroscopic* field. However, the local permittivity profile gives us insight about the influence of the interface. Fig. 4(b) shows that permittivity in the core Si depends only weakly on Si thickness. Its value determines the bulk permittivity in the limit of infinitely thick Si and accordingly we evaluate the dielectric constant of bulk Si to be 11.1, agreeing well with the known value of 11.7. The gradual transition at the interface lowers the permittivity of thin Si but raises the permittivity of the sub-stoichiometric part of the SiO₂, which is in agreement with earlier DFT studies [4], [14].

Finally, we compare the dependence of permittivity on Si thickness, as obtained from our calculations in DFTB for SiO₂- and H-passivated Si. In the latter case, the Si surface is reconstructed by a 1x1 symmetric dihydrate with 1.5 Å Si–H bond-length, consistent with previous studies [3],[4],[6]. The calculations with SiO₂-passivation agree well with the available experimental data from [7], as shown in Fig. 5(a). It is notable however, that H-passivation significantly overestimates the decrease of permittivity with decreasing film thickness. Specifically, we find that for 0.86 nm thick Si film the permittivity is only 17 % lower than our bulk value. In comparison, the decrease in the H-passivated film is 33 %, from our DFTB calculations, and even greater, 35–45 %, is predicted

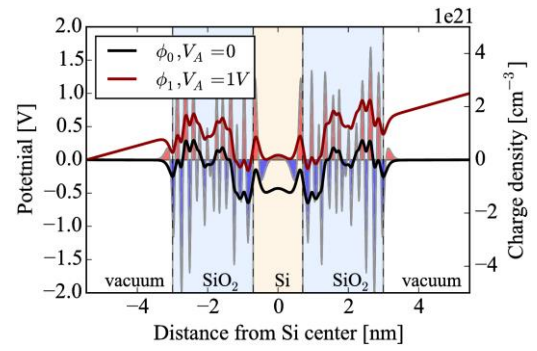


Fig. 2. Potential along the normal of the interface, $\phi(y)$, for zero and one volt applied bias V_A (lines), and charge transfer density, $\delta\rho_0(y)$, at $V_A = 0$ (shaded curves). Quantities are averaged within (xz) -plane; Si film is 1.41 nm.

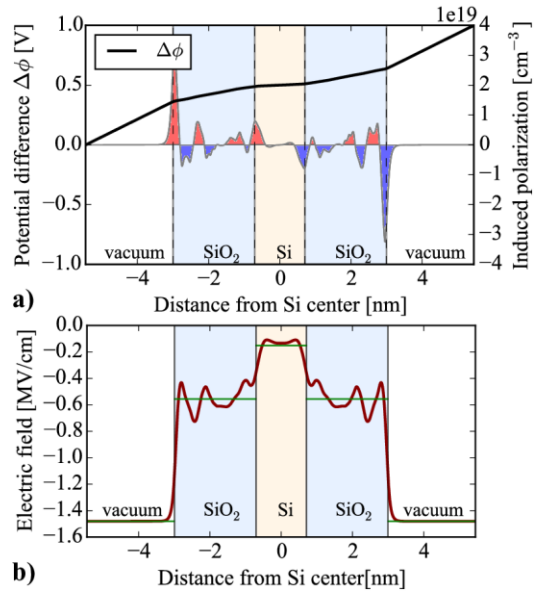


Fig. 3. (a) Difference between the potential with and without applied bias $\Delta\phi(y) = \phi_1(y) - \phi_0(y)$, (line), and the corresponding difference between charge fluctuation densities being the field induced polarization $\rho_{ind} = \delta\rho_1(y) - \delta\rho_0(y)$, (shaded curves). (b) Electric field $E(y) = -\nabla\Delta\phi(y)$. The fluctuations within each layer i are relatively small and averaging $E(y)$ within the layer yields the desired macroscopic field, E_i , depicted as horizontal lines.

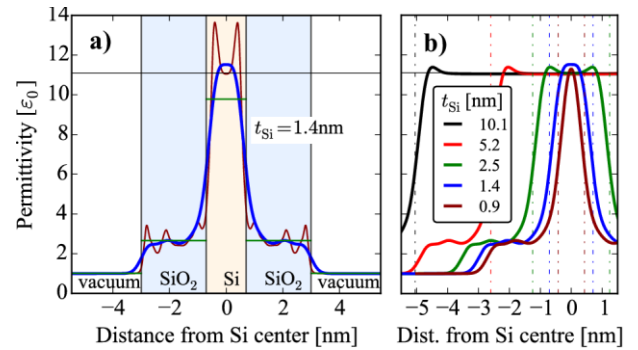


Fig. 4. (a) Microscopic profile of the permittivity $\varepsilon(y)$, obtained from the continuity of the displacement vector at the boundary of each segment of the Poisson grid and taking the electric field $E(y)$ from Fig. 3(b), before (thin line) and after (thick line) Gaussian-kernel smoothing. Average values (note: $1/\varepsilon(y)$ is averaged) shown as horizontal lines. (b). As (a), but for different Si thickness, showing the bulk value arising from the core Si at large thickness.

by DFT calculations with the same atomic model, as shown in Fig. 5(b). We attribute the differences between the results for

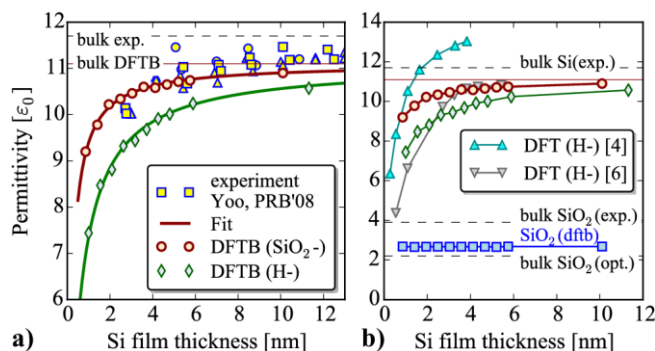


Fig. 5. Permittivity of Si versus its film thickness, calculated with DFTB for SiO₂⁻ and H-passivation is compared against (a)–experiment from [7], and (b)–DFT calculations of H-passivated Si(100) from [4] and [6]. Our calculated value of 2.2 nm SiO₂ permittivity is also shown in (b). Thick lines in (a) are the least-square fit of the analytical model discussed in text.

H-passivated Si to the different methodologies for the evaluation of permittivity, and to the simulations set-up, e.g. Brillouin zone sampling, as detailed in [4]–[6].

It should be emphasized that although the degradation of permittivity in oxidized Si is smaller than previously implied from the hydrogenated models, it is nevertheless significant and very relevant for devices with ultra-thin body. The degradation should be accounted for even in models that use an effective medium to capture the dielectric response of the system. To facilitate that, we have fitted to our numerical results a simple analytical model shown in Fig. 5(a). The model assumes that a Si film with thickness t and permittivity $\kappa(t)\epsilon_0$ is composed of two interfacial layers with thickness d and permittivity $\kappa_i\epsilon_0$, and a Si core layer with thickness $(t - 2d)$ and bulk permittivity $\kappa_b\epsilon_0$. Formally, the governing equation for $\kappa(t)$ becomes: $t/\kappa(t) = 2d/\kappa_i + (t - 2d)/\kappa_b$. We have chosen $d = 0.27$ nm, equivalent to two atomic layers of Si, $\kappa_b = 11.1$, our bulk value discussed above, and fitted κ_i of 8.32 and 5.72 for the oxidized and hydrogenated Si film correspondingly.

We close the discussion with a consideration of the dielectric response in SiO₂. As seen in Fig. 5(b) the result from DFTB is independent of Si thickness and agrees with the optical dielectric constant of SiO₂, which reflects the high-frequency, electronic response. We do not relax the atomic structure under electric field, and do not capture the ionic contribution to permittivity. This is a limitation for atomic systems with ionic bonds, e.g. high- k materials, where ion displacement under electric field forms the main contribution to the large dielectric constant. But it is not intrinsic to the DFTB method – rather, an accurate parameterization of the repulsive interactions of Si and O is missing at present.

In conclusion, the electronic contribution to the dielectric response of Si and SiO₂ is captured intrinsically within DFTB

with good accuracy, in addition to the accurate electronic structure reported in [3]. Therefore, a DFTB Hamiltonian is promising for atomistic modelling of MOSFET, without device- or material-related phenomenological parameters. The simulations of oxidized Si films suggest that the decrease of permittivity due to decreasing Si thickness is unlikely to be a limitation for ultra- and extremely-thin Si-on-insulator devices, as even at 0.9 nm thick Si, the dielectric constant is reduced by less than 18%. The analytical fit reported here enables computationally less demanding models, based on continuum dielectric media, to accurately capture the degradation of Si permittivity as film scaling progresses.

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