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**Citation for published version (APA):**

Schoonus, J. J. H. M., Kohlhepp, J. T., Swagten, H. J. M., & Koopmans, B. (2008). Large magnetoresistance in Si:B-SiO<sub>2</sub>-Al structures. *Journal of Applied Physics*, 103(7, Pt. 3), 07F309-1/3. Article 07F309.  
<https://doi.org/10.1063/1.2832614>

**DOI:**

[10.1063/1.2832614](https://doi.org/10.1063/1.2832614)

**Document status and date:**

Published: 01/01/2008

**Document Version:**

Publisher's PDF, also known as Version of Record (includes final page, issue and volume numbers)

**Please check the document version of this publication:**

- A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher's website.
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## Large magnetoresistance in Si: B-SiO<sub>2</sub>-Al structures

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(Presented on 9 November 2007; received 11 September 2007; accepted 23 October 2007;  
 published online 4 February 2008)

A magnetic-field-dependent resistance change of eight orders of magnitude is observed in boron-doped Si-SiO<sub>2</sub>-Al structures. In order to identify the elementary mechanisms governing this phenomenon, the thickness of the oxidic layer, which is used as an interface energy barrier, has been varied by changing the exposure time to an oxygen plasma. Next, the chemical composition has been monitored by *in situ* x-ray photoelectron spectroscopy measurements. From current-voltage measurements, we observe that at low temperatures, an ultrathin SiO<sub>2</sub> layer provides the kinetic energy to trigger an autocatalytic process of impact ionization. A magnetic field suppresses the onset of impact ionization to higher electric fields, resulting in a large magnetoresistance.

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This paper is focused on the realization and interpretation of extremely large magnetoresistance effects in semiconductors. This magnetoresistance may be utilized for magnetoresistive sensors, which are the critical components in technologies such as high density storage and position/speed monitoring. Silicon is the technologically most common and important semiconductor, and the dependence of the carrier transport on a magnetic field has been investigated frequently.<sup>1,2</sup> A metal-insulator transition,<sup>3</sup> and various other anomalous responses of doped silicon to a magnetic field were reported.<sup>4-7</sup> In line with a reported novel magnetoresistance effect in GaAs,<sup>8</sup> we show at low temperatures in boron-doped Si-SiO<sub>2</sub>-Al structures a robust symmetric positive resistance change of eight orders of magnitude at relatively small magnetic fields of 500 mT. In contrast to other reports on silicon, the magnitude of this resistance change is significantly higher at much smaller magnetic fields, and, moreover, it can be efficiently tuned by the control over the silicon-oxide layer separating the silicon from the nonmagnetic electrodes. In this work, we have varied the thickness of the silicon dioxide layer, and have investigated the chemical composition with x-ray photoelectron spectroscopy (XPS). From transport measurements, we deduce that the origin of this magnetoresistance effect is related to a magnetic-field-controlled process of impact ionization.

We have fabricated a lateral device for electrical characterization, which is based on a (100) boron-doped silicon wafer with a resistivity of 3–9 Ω cm and thickness of 300 μm, supplied by ITME. On top of the native oxide, two aluminum electrodes of 100 μm width at a separation of 50 μm were sputtered. Transport measurements were carried out at 4 K at a bias voltage between 27 and 40 V. As shown in Fig. 1, a magnetic field, aligned parallel to the plane of the substrate and swept from 0 to 500 mT, decreases the current over maximal eight orders of magnitude. We need to point out that the effect is symmetric around zero magnetic field,

and the current was limited up to 10 mA to avoid damage to the device. The corresponding magnetoresistance, defined as  $[R(H)/R(0)-1] \times 100\%$ , with  $R(0)$  and  $R(H)$  the resistance at 10 mA and applied field, respectively, is shown on the right axis. Apart from using a Si:B wafer from ITME, we also prepared devices with identical electrodes and geometry from Si:B wafers supplied by Shin-Etsu (15 Ω cm, 500 μm) and Si-Mat (1–30 Ω cm, 300 μm), which showed reproducible resistance changes of two and eight orders of magnitude, at bias voltages of 20 and 67 V, respectively. These variations could possibly be attributed to the differences in resistivity and mobility of the silicon substrate or a different stoichiometry of the native oxide layer. No pronounced trend in the magnetoresistance is observed when different electrode materials, like tantalum, cobalt, and indium, are used, which proves that the effect is related to intrinsic transport processes in SiSiO<sub>2</sub> structures. We can relate the magnetoresistance directly to the current-voltage behavior, as measured

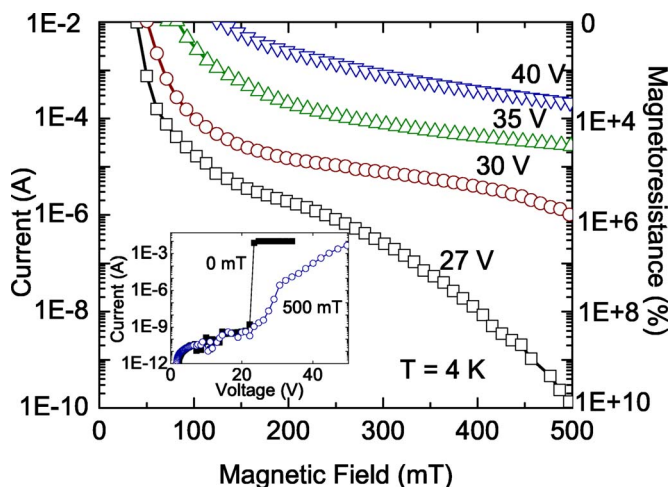


FIG. 1. (Color online) Current as function of the magnetic field (left axis) and corresponding magnetoresistance (right axis), with a current limitation of 10 mA; inset: current-voltage characteristics for 0 and 500 mT, with a current limitation of 10 mA.

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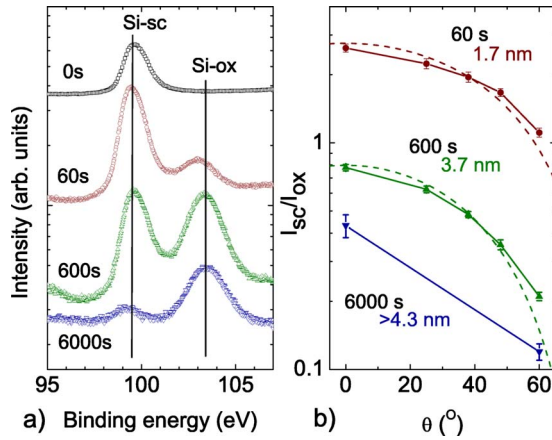


FIG. 2. (Color online) (a) XPS spectra of the variable oxidation of a  $500\ \mu\text{m}$  silicon wafer at an electron exit angle of  $0^\circ$ . (b) Intensity ratios of semiconducting and oxidic silicon as function of the electron exit angle, along with a fit to the Si/SiO<sub>2</sub> overlayer model [Eq. (1)].

for 0 and 500 mT (see inset in Fig. 1). At zero magnetic field at around 25 V, a reversible instantaneous transition to another transport regime occurs. However, no sharp increase is observed in a magnetic field of 500 mT, meaning that the process causing this transition is suppressed.

In order to determine the possible role of the silicon dioxide for these extremely large magnetoresistance effect, we have grown and characterized oxide layers of different thicknesses in controlled environments (base pressure  $< 1 \times 10^{-8}$  mbar). After removal of the native surface oxide layer with a  $100\ \text{mol/m}^3$  HF dip, clean oxide layers were prepared on the silicon (Shin-etsu  $15\ \Omega\ \text{cm}$ ,  $500\ \mu\text{m}$ , dopant boron) by plasma oxidation at a pressure of  $1 \times 10^{-1}$  mbar at 15 W for 0, 60, 600, and 6000 s.<sup>9</sup> To identify the chemical composition of the surface, *in situ* XPS (Mg  $K\alpha$ , 1253.6 eV) has been used. Silicon in an oxide surrounding and semiconducting crystalline silicon have binding energies of 103.4 and 99.2 eV, respectively, as shown in the Si  $2p$  spectra in Fig. 2(a) for an electron exit angle  $\Theta=0^\circ$  with respect to the surface normal. The total peak intensities are related to the stoichiometry of the surface. Directly after the HF dip, no oxygen is present. For increasing oxidation times, the relative amount of silicon dioxide increases at the cost of the semiconducting silicon. The oxidic portion of the Si  $2p$  spectra shows a small binding energy shift for larger oxidation times, indicating that the chemical nature of the oxide is still changing, and complete stoichiometry is not reached for incomplete oxidized silicon.

For an estimation of the oxide thickness  $d_{ox}$ , we have measured the ratio of oxidic and semiconducting silicon total peak intensities  $I_{sc}/I_{ox}$  for different electron exit angles [Fig. 2(b)].  $I_{sc}/I_{ox}$  can be calculated by a simple Si/SiO<sub>2</sub> overlayer model, which is based for each layer on the sum over all atoms at all depths, and corrects for the escape depth of the electrons:

$$\frac{I_{sc}}{I_{ox}} = \frac{\lambda_{sc} c_{sc} e^{[d_{ox}(c_{ox}/c_{sc})-d_0]} - 1}{\lambda_{ox} c_{ox} 1 - e^{d_{ox}[\lambda_{ox} \cos \Theta]}} \quad (1)$$

with  $c_{ox}/c_{sc}=2.9 \pm 0.3$  the ratio of the atomic concentrations (which may be not fully justified for the device with 60 s

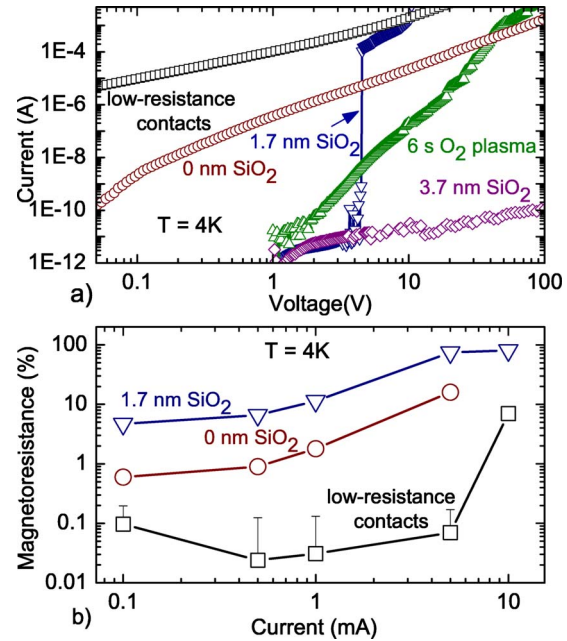


FIG. 3. (Color online) (a) Current-voltage characteristics for different metal/silicon contacts and electrode spacing of 0.5 mm. (b) Magnetoresistance taken at constant currents measured at 502 mT.

plasma oxidation),  $d_0$  the thickness of the sample, and the escape depths of the electrons in silicon and SiO<sub>2</sub> being  $\lambda_{sc}=2.0\ \text{nm}$  and  $\lambda_{ox}=2.3\ \text{nm}$ , respectively.<sup>10</sup> For 60 and 600 s plasma oxidation, the measured intensity ratios have been fitted to the overlayer model, and we obtain oxide thicknesses of 1.7 and 3.7 nm, respectively. For 6000 s plasma oxidation,  $I_{sc}/I_{ox}$  is only measured for  $\Theta=0^\circ$  and  $60^\circ$ , and cannot be accurately fitted. Nevertheless, based on the value of  $I_{sc}/I_{ox}$  at  $\Theta=0^\circ$  and Eq. (1), the thickness is estimated to be larger than 4.3 nm.

Subsequently, we want to establish the combined influence of the applied magnetic field and the energy barrier of the electrode-silicon contact on the electrical transport characteristics. Therefore, a low-resistance contact Si:B–Al device and Si:B–SiO<sub>2</sub>–Al devices are prepared with silicon dioxide layers formed by 0 s (0 nm), 6 s (no thickness measured), 60 s (1.7 nm), 600 s (3.7 nm), and 6000 s ( $>4.3$  nm) oxygen plasma. Contrary to the experiments with the native oxide, for these experiments, the electrodes are 1 mm wide, separated at 0.5 mm, and have been grown *in situ* by dc magnetron sputtering. The low-resistance contacts have been prepared by annealing a sample with 0 nm SiO<sub>2</sub> in argon atmosphere at  $450^\circ\ \text{C}$  for 30 min. No annealing step was used on all other devices to avoid diffusion of impurities into the oxide layer and silicon substrate, thereby ensuring clearly distinctive interface barriers.

For the Si:B–(SiO<sub>2</sub>)–Al structures, the  $I$ - $V$  characteristics have been measured at 4 K [Fig. 3(a)]. Although the exact line shapes are difficult to explain in detail, we observe, as expected, that for low bias voltages the total device resistance increases from the low resistive contacts towards contacts with a thick oxide, corresponding to higher and wider interface barriers. Surprisingly, for higher bias voltages, the insertion of an extra interface resistance can lead to

a lower total device resistance compared to a device without this extra interface resistance, which is seen for the devices with a 6 s plasma oxidation and 1.7 nm SiO<sub>2</sub>. This behavior can be explained by a dramatic suppression of the silicon resistance due to an autocatalytic process of impact ionization of the shallow acceptor boron.<sup>8,11,12</sup> Therefore, we conclude that an ultrathin interface barrier assists the autocatalytic process of impact ionization, and speculate that this process is triggered by hot holes which gain by tunneling sufficient kinetic energy from the voltage drop over the interface barrier to ionize neutral boron atoms.<sup>13</sup> To sustain the breakdown, the electric field in the silicon should be large enough to increase within the mean free path the kinetic energy of the holes to what is required for another impact ionization. For a device with a SiO<sub>2</sub> layer of 1.7 nm, the impact ionization starts around 4 V. The bulk conductivity increases, thereby the voltage over the oxide layer increases, by which the process of impact ionization is accelerated, finally causing an almost immediate increase in current. For a device with a SiO<sub>2</sub> layer of 3.7 nm, a breakdown is likely to be expected as well, although for bias voltages higher than 100 V.

Although the magnetoresistance measured at constant current is always smaller than the constant voltage magnetoresistance, in this configuration, the voltage drop over the interface barrier is independent of the magnetic field, which enables us to determine accurately the influence of the energy barrier on the magnetoresistance. As expected, the devices with >4.3 nm and 3.7 nm tunnel barriers of SiO<sub>2</sub> are too resistive for further magnetoresistive analysis. The device with 6 s plasma oxidated silicon does not have a closed oxide layer, resulting in a nonhomogeneous current flow, and magnetoresistance data are not conclusive. The constant current magnetoresistance of the device with a 1.7 nm tunnel barrier of SiO<sub>2</sub> is largest for any measured current and increases with current [Fig. 3(b)]. With just a Schottky barrier (0 nm SiO<sub>2</sub>), the magnetoresistance for all currents is a factor of 10 lower than the device with a 1.7 nm SiO<sub>2</sub>, whereas with low-resistance contacts, the magnetoresistance is negligibly small (maximum 7% at 10 mA) and no pronounced trend is visible.

We now turn to the underlying mechanism that could explain the magnetoresistance. As suggested by Sladek<sup>14</sup> for *n*-InSb, at small enough impurity concentrations, the acceptor wave functions are centered on the acceptor atoms, but they still have a finite overlap. The magnetic field causes shrinkage of the acceptor wave functions. The hole orbitals become more localized in the vicinity of the acceptor ions, the overlap by the tails is reduced, and the magnetic field gradually narrows the previously spread-out acceptor levels into an impurity band, which resides at a higher energy. We have deduced from admittance spectroscopy measurements that a magnetic field of 500 mT raises the effective acceptor level by 1.8 meV compared to the valence band, assuming the rate of single hole capture is independent of the field.<sup>15</sup> For acceptor levels with an energy large compared to *kT*, the

probability of impact ionization exponentially decreases with the depth of the acceptor level, and exponentially increases with the hole velocity.<sup>16</sup> Additionally, the recombination process is less effective when holes have high velocity since the capture cross section decreases with increasing carrier energy.

In view of these considerations, Fig. 3(b) can be understood as follows. For small hole velocities (i.e., less wide energy barrier or low current), the rate of single hole capture is a few orders of magnitude larger than the impact ionization,<sup>16,17</sup> and most carriers are in the impurity band with low mobility [see bottom left, Fig. 3(b)]. To obtain a large magnetoresistance, two conditions should be fulfilled. First, because the magnetoresistance is proportional to  $V_{Si}/V_{total}$ , the largest voltage drop has to be over the silicon. Consistent with this, we have measured that the magnetoresistance effect increases with electrode spacing.<sup>15</sup> Second, the voltage drop over the oxide must be large enough to supply the kinetic energy to the carriers required for impact ionization process, which is subject to the acceptor energy and, thus, the magnetic field. This condition is fulfilled by either increasing the current or increasing the oxide thickness [Fig. 3(b)], by which the voltage over the oxide and the magnetoresistance increases.

In summary, we observe a positive resistance change of eight orders of magnitude at magnetic fields of 500 mT in boron-doped Si-native SiO<sub>2</sub>-Al structures. From devices with SiO<sub>2</sub> layers of various thicknesses, which are grown in a controlled environment and characterized by XPS, we conclude that an ultrathin layer of SiO<sub>2</sub> assists the magnetic-field-controlled process of impact ionization.

This work was supported by the Dutch Technology Foundation (STW) via the NWO VICI-grant "Spintronics."

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