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Field dependent permittivity in metal-semiconducting SrTiO₃ Schottky diodes

R. A. van der Berg,^{a)} P. W. M. Blom, J. F. M. Cillessen, and R. M. Wolf
Philips Research Laboratories, Prof. Holstlaan 4, 5656 AA Eindhoven, The Netherlands

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The field dependence of the dielectric constant of SrTiO₃ is investigated using the capacitance versus bias voltage characteristics of various metal-SrTiO₃:Nb Schottky diodes. The relative dielectric constant is shown to decrease one order of magnitude for electric fields ranging from 0.1 to 10 MV/cm. At low fields the permittivity follows the Curie–Weiss law, whereas at fields larger than 500 kV/cm the permittivity is nearly temperature independent. At high doping densities the field dependent permittivity gives rise to a reduction of the depletion width of the Schottky diode. © 1995 American Institute of Physics.

Strontium titanate (SrTiO₃) single crystals are widely used as a substrate for epitaxial growth of ferroelectrics such as lead- and barium titanate, ferromagnetics such as lanthanum strontium manganate, and superconductors such as yttrium barium copper oxide. Furthermore, because of its high dielectric constant ϵ_r (≈ 300) insulating SrTiO₃ single crystals are also used as an active gate in dielectric-base tunnel transistors.¹ SrTiO₃ is also attractive as dielectric in thin-film capacitors, not only due to its high permittivity but also because of the low leakage current densities. In view of these applications the question arises whether the high permittivity of SrTiO₃ is affected by a large electric field, since in thin films high electric fields are produced even at low applied voltages. The permittivity of single-crystal SrTiO₃ was studied by Neville *et al.*,² who reported that above 60 K the relative permittivity is characterized by a Curie–Weiss law with a Curie temperature of 30 K. Also, for temperatures below 65 K a field dependent permittivity was found, which was attributed to the hardening of the motion of the titanium ions. In this study, only electric fields up to 23 kV/cm were applied and due to these low fields no field dependent permittivity for temperatures above 65 K was observed.

The application of Schottky barriers enables the determination of the dielectric constant at electric fields which are not addressable by other methods. The field dependence of the dielectric constant modifies the capacitance–voltage (C – V) characteristics of a Schottky barrier.³ From the capacitance data of a gold-(reduced) potassium tantalate (KTaO_{3-x}) Schottky diode a strong decrease of the permittivity with increasing interface field was obtained. Differential capacitance results for Schottky barriers formed between indium and reduced SrTiO₃ (SrTiO_{3-x}) at room temperature by Hayashi and Aoki,⁴ however, are well explained by a field independent dielectric constant. Only at low temperatures (<4.2 K) a discrepancy between the experimental and calculated results appears. This discrepancy is attributed to the electric field dependence of the dielectric constant of SrTiO₃ at low temperature. However, as a result of the low carrier concentration ($<4 \times 10^{18}$ cm⁻³) and the small maximum re-

verse bias (<-4 V) applied in this study, the maximum electric fields obtained at the interface of the In-SrTiO_{3-x} Schottky diodes are relatively low (<250 kV/cm). Finally, in tunneling measurements at $T=4.2$ K by Sroubek⁵ through niobium doped SrTiO₃–indium (SrTiO₃:Nb–In) Schottky barriers an effective dielectric constant of about 10 is used in order to obtain reasonable barrier widths. This reduction of the dielectric constant is partly attributed to the high electric field present in the depletion layer of the Schottky diode. Thus, the dielectric constants of SrTiO₃ reported so far have only been measured at relatively low fields and no dependence on the electric field has been observed at room temperature. In the present study the field dependent permittivity of SrTiO₃ up to very high fields is obtained from capacitance voltage measurements on mercury-(Hg) and gold (Au)-SrTiO₃:Nb Schottky diodes. By varying the Nb-donor density the electron concentration in the various SrTiO₃:Nb single crystals ranges from 2×10^{18} to 1×10^{20} cm⁻³. As a result we are able to obtain a permittivity profile for interface fields ranging from 200 kV/cm to 10 MV/cm. We observe that for electric fields larger than 300 kV/cm the relative dielectric constant starts to decrease gradually from 300 to 25 at 10 MV/cm. From our temperature dependent capacitance voltage measurements we obtain that at low fields the measured permittivity is in agreement with the predictions of the Curie–Weiss law.² For electric fields larger than 500 kV/cm the permittivity is temperature independent. The field dependence of the dielectric constant is shown to be relevant for the spatial distribution of the built-in electric field and the resulting depletion width in a Schottky diode.

In our experiments we use Verneuil grown Nb-doped SrTiO₃ crystals, which are obtained in boules with a maximum diameter of about 15 mm and cut into 1 mm thick slices in the (100) direction. One side of the sample is Syton polished. The Nb content as measured by x-ray fluorescence (XRF) and laser ablated inductively coupled mass spectrometry (LA-ICP-MS) amounts to 160, 940, and 2750 wt. ppm, respectively. From Hall measurements the corresponding electron concentrations of 2.5×10^{18} , 2.3×10^{19} , and 1.0×10^{20} cm⁻³ are obtained, respectively. Schottky contacts are fabricated on the polished side of the samples using evaporated Au contacts as well as a mercury (Hg) probe.

^{a)}Present address: Department of Applied Physics and Materials Science Center, University of Groningen, Nijenborgh 4, 9747 AG Groningen, The Netherlands. Electronic mail: blom@prl.philips.nl

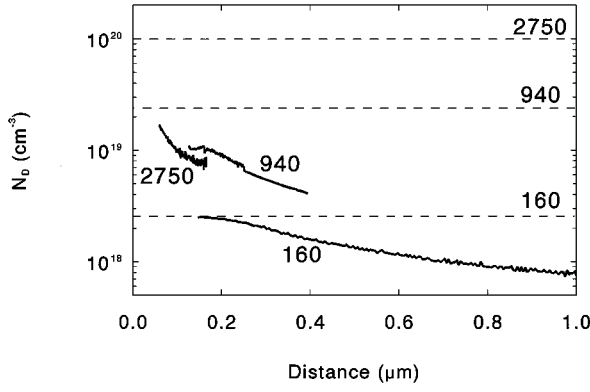


FIG. 1. Dope profiles measured with Schottky barriers on various SrTiO₃:Nb single crystals (solid lines) assuming a constant relative dielectric constant of $\epsilon_r=300$. Also shown are the dope profiles as expected from Hall measurements (dashed lines) for Nb contents of 160, 940, and 2750 wt. ppm, which correspond to electron densities of 2.5×10^{18} , 2.3×10^{19} , and $1 \times 10^{20} \text{ cm}^{-3}$, respectively. The measured profiles decrease with increasing depletion depth and are all below the carrier densities as obtained from Hall measurements. This discrepancy increases with increasing dopant density.

Ohmic contacts on the bottom side are made by ultrasonic soldering or using Ga/In eutect. For a field independent dielectric constant the doping profile can be directly obtained from the $C-V$ measurements, since

$$N_D = \frac{2}{q\epsilon} \left(-\frac{1}{d(1/C^2)/dV} \right) \quad (1)$$

with N_D the donor density at the space charge edge, ϵ the dielectric constant, C the diode capacitance per area, and V the reverse bias voltage. We expect to see a constant doping profile because SIMS measurements show a homogeneous Nb distribution as a function of depth. In Fig. 1 the dope profiles measured with the Hg diode are shown. We observe an apparent decreasing donor concentration with increasing depletion depth for all crystals. A similar behavior was observed for the Au-SrTiO₃:Nb diodes. Furthermore, the profiles of the Nb doped crystals are all below the value of the measured (Hall) carrier density and the discrepancy increases with increasing dopant density. For the highest dopant density (2750 ppm) the difference between the expected ($1 \times 10^{20} \text{ cm}^{-3}$) and measured ($1.7 \times 10^{19} \text{ cm}^{-3}$) dopant density almost amounts to one order of magnitude. However, a field dependence of ϵ gives rise to appreciable deviations of the standard Schottky theory, as we will now show. For a field dependent dielectric constant ϵ is defined as³

$$\epsilon(x) = \frac{dD(x)}{dE(x)}, \quad (2)$$

with D the dielectric displacement and E the electric field. The capacitance per area is then given by

$$\frac{1}{C} = \int_0^W \frac{dx}{\epsilon(x)}, \quad (3)$$

with $x=0$ at the metal semiconductor interface and $x=W$ at the space charge edge. Assuming now, a uniform charge distribution the electric field at $x=0$ and the corresponding dielectric constant are obtained from

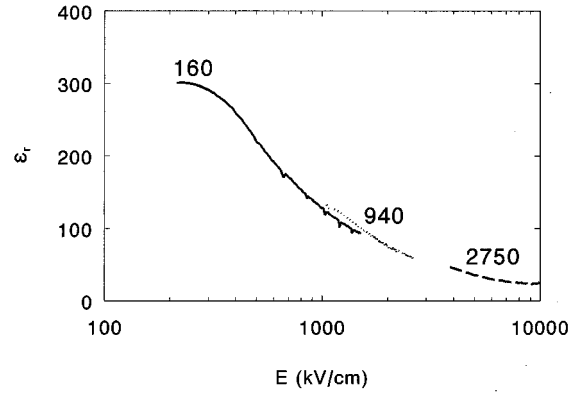


FIG. 2. Permittivity profile of SrTiO₃ at room temperature as determined from $C-V$ relations of the rectifying contact of Hg on homogeneously Nb doped SrTiO₃ with Nb contents of 160, 940, and 2750 wt. ppm. For fields larger than 300 kV/cm the relative permittivity decreases from 300 to a minimum of 25 at a field of 10 MV/cm.

$$E(0) = - \left(\frac{qN_D}{C} \right) \quad (4)$$

and

$$- \frac{\partial}{\partial V} (1/C^2) = \frac{2}{qN_D \epsilon(0)}. \quad (5)$$

Equations (4) and (5) form the equivalent of the dope profile measurements, which assume a field independent permittivity but an arbitrary dope profile. Applying Eqs. (4) and (5) to our Hg diode data of Fig. 1 we observe that the permittivity profiles for the Nb doped SrTiO₃ crystals all coincide on a single curve as shown in Fig. 2. This is strong evidence that SrTiO₃ has a field dependent permittivity which reveals itself at high electric fields. At fields lower than 200 kV/cm the relative dielectric constant amounts to 300, which is in agreement with the earlier reported values.² This also explains the absence of a field dependent ϵ in earlier measurements^{2,4} on SrTiO₃ because the applied electric fields were too low.

A further evidence for the occurrence of a field dependent ϵ comes from the measurement of the permittivity profiles as a function of temperature, which then are compared with the Curie-Weiss law given by²

$$\epsilon(T) = \frac{A}{T - T_c} \epsilon_0, \quad (6)$$

with $A=8.9 \times 10^4 \text{ K}$ and $T_c=30 \text{ K}$. In Fig. 3 the temperature dependence of the relative permittivity is shown from 140 to 400 K for the low doped (160 wt. ppm) sample, furthermore, the permittivity as expected from the Curie-Weiss law [Eq. (6)] is indicated. The permittivity increases with decreasing temperature and is in excellent agreement with the predictions of the Curie-Weiss law at low fields. The lower permittivities obtained at high electric fields show no strong temperature dependence, at fields higher than 500 kV/cm the permittivity is even independent of temperature.

The positional dependence of the internal field in the space charge region of a Schottky diode implies that the dielectric constant of the semiconducting SrTiO₃ will also be

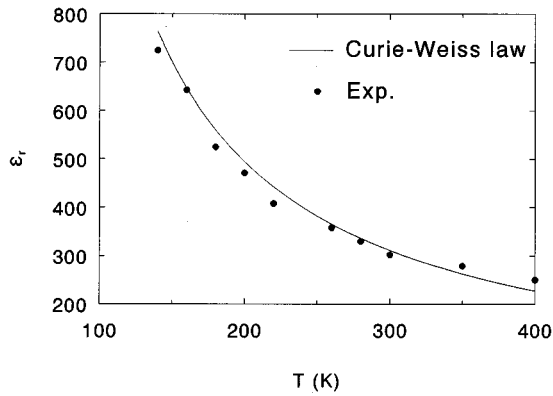


FIG. 3. Relative permittivity of SrTiO₃ as a function of temperature. At low fields the permittivity follows the Curie–Weiss law [Eq. (6)] with $A=8.4\times 10^4$ K and $T_c=30$ K. For fields larger than 500 kV/cm no temperature dependence of the permittivity is observed.

a function of the position. The consequences of the field dependent permittivity for the band bending of a metal-semiconducting SrTiO₃ Schottky diode are schematically depicted in Fig. 4. The positive direction of the electric field is defined as pointing from the metal into the semiconductor. At the metal-semiconductor interface, where the built-in electric field reaches its maximum value, the dielectric constant is reduced when the built-in field exceeds 300 kV/cm. The reduction of the dielectric constant in the high-field part of the space charge region then gives rise to a decrease of the depletion width. In Fig. 4 the calculated energy-band diagram [Fig. 4(a)], the electric field [Fig. 4(b)], and dielectric constant [Fig. 4(c)] are shown as a function of distance for constant ϵ_r (300) and $\epsilon_r(E)$, as determined from Fig. 2. In our calculation a barrier height of 1.2 eV,^{6,7} a built-in voltage of 1.1 eV, and a carrier concentration of 2×10^{19} cm⁻³ are used. For the field dependent ϵ_r , a depletion width of 35.7 nm is obtained, whereas $\epsilon_r=300$ gives rise to a depletion width of 42.7 nm. The difference in capacitance of the space charge region, 1.88 and 1.95 nF, respectively, is smaller because the difference in depletion width is partly compensated by the different dielectric constants. It is evident that for low doped semiconductors, where the internal field at the interface is smaller than 300 kV/cm, the effect of a field dependent dielectric constant is strongly reduced.

In conclusion, we have measured the field dependence of the permittivity of SrTiO₃ at room temperature. The electric field ranges from 200 kV/cm up to the very large value of 10 MV/cm by using the built-in field of Schottky diodes fabricated on semiconducting SrTiO₃:Nb with various dopant densities. For electric fields larger than 300 kV/cm the per-

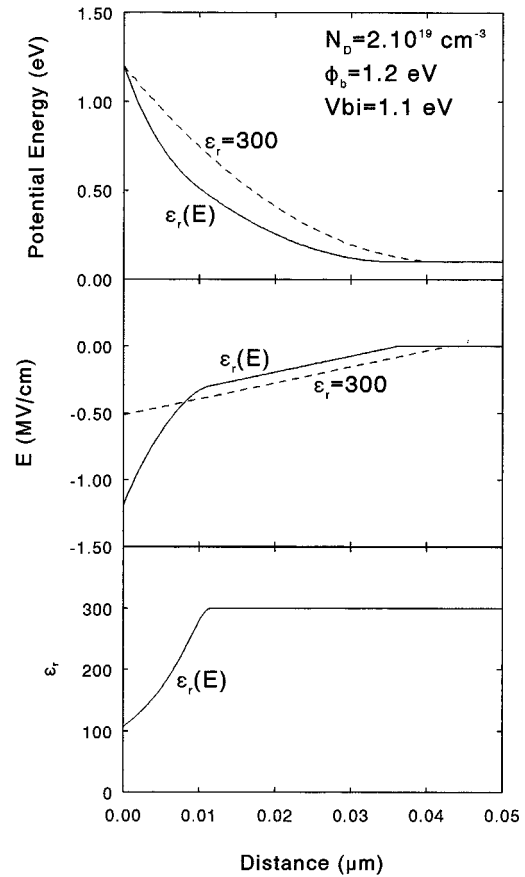


FIG. 4. Calculated energy band diagram (a), electric field (b), and relative dielectric constant (c) as a function of position for a metal-SrTiO₃:Nb Schottky contact. The decrease of the dielectric constant (c) as a result of the high fields (>300 kV/cm), which are present close to the metal-semiconductor interface (b), gives rise to a decrease of the depletion width (a).

mittivity gradually decreases from 300 to 25 at 10 MV/cm. At low fields, the permittivity is governed by the Curie–Weiss law in a temperature range from 140 to 400 K. At fields larger than 500 kV/cm the permittivity is nearly temperature independent. For Schottky barriers on highly doped SrTiO₃ the field dependent permittivity gives rise to a considerable reduction of the depletion width.

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