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**LETTER TO THE EDITOR**

# Negative differential resistivity and positive temperature coefficient of resistivity effect in the diffusion-limited current of ferroelectric thin-film capacitors

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**Abstract**
We present a model for the leakage current in ferroelectric thin-film capacitors which explains two of the observed phenomena that have escaped satisfactory explanation, i.e. the occurrence of either a plateau or negative differential resistivity at low voltages, and the observation of a positive temperature coefficient of resistivity (PTCR) effect in certain samples in the high-voltage regime. The leakage current is modelled by considering a diffusion-limited current process, which in the high-voltage regime recovers the diffusion-limited Schottky relationship of Simmons already shown to be applicable in these systems.

For a number of years the problem of understanding leakage currents in ferroelectric capacitors (or high-dielectric-constant capacitors using ferroelectric materials just above their phase transtion, e.g. barium strontium titanate (BST)) has been of practical interest, and a large number of papers have been published on the subject, a review of which can be found in Scott's book [1]. Typically, the currents observed appear to be either emission limited or space charge limited. In this paper we restrict ourselves to the discussion of the emission-limited type currents. What this paper shows is that a metal–ferroelectric–metal system with space charge due to oxygen vacancies can be considered as a diffusion-limited current system. The diffusion-limited current has two regimes: a low-voltage regime akin to the low-voltage regime discussed in the theory of semiconductor punch-through diodes [2]; and a high-voltage regime equivalent to the diffusion-limited Schottky relationship of Simmons [3].

We consider the field distribution in a capacitor (figure 1) which, as well as having a large dielectric constant $\varepsilon_s$ and high uniform concentration of stationary oxygen vacancies

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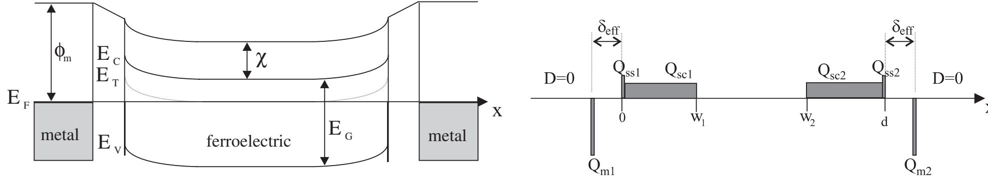


**Figure 1.** Band diagram and representation of charges used to calculate field and potential distributions.

$N_D$, has a remnant polarization, $P_r$. Although the oxygen vacancy distribution is considered constant throughout the film the vacancies contribute space charge only when they lie above the Fermi level and are unoccupied by electrons, i.e. in the depletion widths $w_1$ and $w_2$. The Thomas–Fermi screening length in the electrode and the penetration of the metal states into the ferroelectric are accounted for by separating $Q_m$, the charge on the metal from $Q_{ss}$, the charge due to surface states (metal-induced gap states) by an effective thickness $\delta_{eff}$. This effective thickness reflects the fact that there is a finite screening length in the metal (the Thomas–Fermi screening length $\lambda_{tf}$) and that metal-induced gap states penetrate a small distance into the insulator ($\lambda_{MIGS}$).

$$\delta_{eff} = \frac{\lambda_{tf}}{\varepsilon_0} + \frac{\lambda_{MIGS}}{\varepsilon_\infty \varepsilon_0}. \tag{1}$$

Metal-induced gap states have previously have been shown to be important in ferroelectric thin-film capacitors by Robertson and Chen [4]. The metal screening length term for a good metal is around 0.5 Å whilst *ab initio* simulations of the Pt–$BaTiO_3$ interface by Rao *et al* [5] suggest a MIG penetration length $\lambda_{MIGS}$ of about 1.7 Å and $\varepsilon_\infty$ is around 5.6, so $\delta_{eff}$ is usually between about 0.5 and 1 Å$/\varepsilon_0$. In the following equations $q$ is the charge of an electron.

We now consider that the electric displacement in the film is given by a field-independent component, the remnant polarization $P_r$ and a linear dielectric component $\varepsilon_s \varepsilon_0 E$ where the static dielectric constant of the film is $\varepsilon_s$ and $E$ is the electric field,

$$D = \varepsilon_s \varepsilon_0 E \pm P_r. \tag{2}$$

The sum of the charges in the system must equal zero since the electric displacement is zero in both electrodes (far from the interface). In the centre of the film the field must equal the field that would arise if there were no surface states or space charge, which we take to be the linear drop ($\frac{\partial V}{\partial x} = \frac{V}{d}$) that would occur if the sample were a perfect insulator. This is quite different from the approach often taken, which is to treat a metal–semiconductor–metal system as two semi-infinite junctions, with the electric field taken as zero in the regions of the film where there is no space charge, regardless of the applied field. Such an approach would be incompatible with the space-charge-free limit of a linear voltage drop for our insulating system. (This approach is however perfectly appropriate for a single infinite-metal–infinite-semiconductor junction.) In addition, because our system is ferroelectric, when there is a spontaneous polarization a finite potential drop across the system occurs because of the imperfect screening of the depolarization field, even in the absence of an applied field. Using these arguments one finds that the field in the space-charge-free interior of the film is

$$E_0 = -\frac{1}{\varepsilon_s \varepsilon_0} \frac{V \pm P_r 2\delta_{eff}}{2\delta_{eff} + \frac{d}{\varepsilon_s \varepsilon_0}}. \tag{3}$$

Our expressions for the charge on the metal electrodes ($Q_{m1}$ and $Q_{m2}$) are then

$$Q_{m1} = -Q_{ss1} - qN_D w_1 - \frac{V \mp P_r \frac{d}{\varepsilon_s \varepsilon_0}}{2\delta_{eff} + \frac{d}{\varepsilon_s \varepsilon_0}} \tag{4}$$

**Table 1.** Expressions for the potentials and fields in the different regions of the capacitor.

| Region | Potential | Field |
|---|---|---|
| $0 \leqslant x \leqslant w_1$ | $Q_{m1}\delta_{eff} - E_0\frac{x}{\varepsilon_s\varepsilon_0} + \frac{qN_D}{\varepsilon_s\varepsilon_0}x\left(w_1 - \frac{x}{2}\right)$ | $E(x) = E_0 - \frac{qN_D}{\varepsilon_s\varepsilon_0}(w_1 - x)$ |
| $w_1 < x < d - w_2$ | $Q_{m1}\delta_{eff} - E_0\frac{x}{\varepsilon_s\varepsilon_0} + \frac{qN_D}{2\varepsilon_s\varepsilon_0}w_1^2$ | $E(x) = E_0$ |
| $d - w_2 < x < d$ | $Q_{m1}\delta_{eff} - E_0\frac{x}{\varepsilon_s\varepsilon_0} + \frac{qN_D}{2\varepsilon_s\varepsilon_0}w_1^2 - \frac{qN_D}{2\varepsilon_s\varepsilon_0}(x - (d - w_2))^2$ | $E(x) = E_0 + \frac{qN_D}{\varepsilon_s\varepsilon_0}(x - (d - w_2))$ |

$$Q_{m2} = -Q_{ss2} - qN_D w_2 + \frac{V \mp P_r\frac{d}{\varepsilon_s\varepsilon_0}}{2\delta_{eff} + \frac{d}{\varepsilon_s\varepsilon_0}}. \tag{5}$$

The amount of charge due to surface states ($Q_{ss1}$ and $Q_{ss2}$) is independent of voltage and is found by the condition that the Fermi level at the interface of the ferroelectric is $S(\phi_m) + (1 - S)(\chi + (E_c - \phi))$ below the vacuum level where $S = \frac{1}{1+q^2\delta_{eff}D_s}$ and $\phi$ is the charge neutrality level (following Robertson and Chen [4] who give a value of $S$ for $BaTiO_3$ of 0.28); this gives

$$Q_{ss1} = (1 - S)\left(-qN_D w_1 + \frac{1}{\delta_{eff}}((\phi_m - \chi) - (E_g - \phi))\right). \tag{6}$$

This result shows that the charge due to surface states is independent of voltage, and under the assumptions that the two electrodes are the same and that the film is uniform, $Q_{ss1} = Q_{ss2}$.

Finally, the width of the depletion layers is found using the condition that the potential at $x = w_1$ must be equal to the height of the donor trap level $E_t$ (which in samples with low intrinsic conductivity and high dopant levels will be the semiconductor Fermi level). It is found that the depletion regions are invariant with applied voltage,

$$w_1 + \varepsilon_s\varepsilon_0 S\delta_{eff} = \left[(\varepsilon_s\varepsilon_0 S\delta_{eff})^2 + \frac{2\varepsilon_s\varepsilon_0}{qN_D}((\phi_m - \chi) - (E_c - E_t) - (1 - S)((\phi_m - \chi) - (E_c - \phi)))\right]^{\frac{1}{2}} \tag{7}$$

and that for a symmetrical system $w_2 = w_1$. If a film is fully depleted (i.e., if the calculated values of $w_1$ and $w_2$ are greater than $\frac{d}{2}$ (which means that the system Fermi level never reaches the trap level)), the following equations may be used with $w_1 = w_2 = \frac{d}{2}$.

The finding that the depletion widths do not change with field is at first quite surprising, the typical idea being that in punch-through diodes depletion widths will change with field and that at a certain applied bias the reverse-biased depletion width will 'punch through' to the forward-biased Schottky barrier. But in fact what we show here is that it is not the depletion width itself that changes size, but rather the physical location within the system of the field inversion point. It is when this field inversion point coincides with the forward-biased Schottky barrier that punch-through behaviour occurs. The expressions for the potentials and fields in the different regions of the capacitor are displayed in table 1. An important result of this system of equations is that below a certain critical voltage there is a point

$$x_0 = -\frac{\varepsilon_s\varepsilon_0}{qN_D}E_0 + d - w_2 \tag{8}$$

in the film at which the field is zero (the field inversion point referred to above). We can calculate the current flowing in the device by determining the point in the film at which the

field is zero. Here the current is entirely due to diffusion and is given by

$$J = q\mu kT\frac{\partial n}{\partial x}. \qquad (9)$$

To evaluate $\frac{\partial n}{\partial x}$ we look at the number of conduction electrons in the region $d-w_2 < x < d$, which (under the assumption that all the conduction electrons come from the oxygen vacancy trap level) is given by

$$n(x) = N_{\mathrm{D}}\exp\left(-\frac{q}{kT}\left((E_{\mathrm{c}}-E_{\mathrm{t}}) + \frac{qN_{\mathrm{D}}}{2\varepsilon_{\mathrm{s}}\varepsilon_0}(x-(d-w_2))^2\right)\right). \qquad (10)$$

Taking the derivative and using the solution for $x_0$ from above we find the current to be

$$J = q\mu N_{\mathrm{D}}E_0\exp\left(-\frac{q}{kT}\left((E_{\mathrm{c}}-E_{\mathrm{t}}) + \frac{\varepsilon_{\mathrm{s}}\varepsilon_0}{2qN_{\mathrm{D}}}(E_0)^2\right)\right). \qquad (11)$$

An equivalent expression for the current, which is also applicable for fully depleted films (in contrast to the earlier expression, which applies to only partially depleted films), is

$$J = q\mu N_{\mathrm{D}}E_0\exp\left(-\frac{q}{kT}\left(\phi_{\mathrm{b}} - \frac{qN_{\mathrm{D}}w^2}{2\varepsilon_{\mathrm{s}}\varepsilon_0} + \frac{\varepsilon_{\mathrm{s}}\varepsilon_0}{2qN_{\mathrm{D}}}(E_0)^2\right)\right). \qquad (12)$$

For a fully depleted film one simply substitutes $w = \frac{d}{2}$ into the equation above. These relationships are applicable until $x_0 = d$ which is when $\frac{V}{2\delta_{\mathrm{eff}}\varepsilon_{\mathrm{s}}\varepsilon_0+d} = qN_{\mathrm{D}}w_2$ (or in other words when the field at the cathode is zero). This is equivalent to punch-through, but a punch-through of the field inversion point, rather than the edge of the depletion width; and it marks the transition to a new conduction regime. The current at which the transition occurs is the same as Frank and Simmons obtain [6], i.e.,

$$J_{\mathrm{T}} = -q\mu N_{\mathrm{D}}\frac{qN_{\mathrm{D}}w_2}{\varepsilon_{\mathrm{s}}\varepsilon_0}\exp\left(-\frac{\phi_{\mathrm{b}}}{kT}\right). \qquad (13)$$

The voltage at which the transition occurs,

$$V = qN_{\mathrm{D}}w_2\left(2\delta_{\mathrm{eff}} + \frac{d}{\varepsilon_{\mathrm{s}}\varepsilon_0}\right) \qquad (14)$$

is temperature and thickness dependent; the temperature dependence is somewhat complicated because of the temperature dependence of the dielectric constant (and hence also the depletion width). When the applied voltage is increased beyond this voltage, the image force pushes the potential maximum back into the film, lowering the effective barrier. If we assume that the new potential maximum is quite close to the interface, we can use the field at the interface to calculate the Schottky barrier lowering as

$$\Delta\phi_{\mathrm{b}} = \sqrt{\frac{q}{4\pi\varepsilon_\infty\varepsilon_0}\left(-E_0 - \frac{qN_{\mathrm{D}}w_2}{\varepsilon_{\mathrm{s}}\varepsilon_0}\right)}. \qquad (15)$$

The dielectric constant $\varepsilon_\infty$ used here is the optical dielectric constant of the material ($\approx$5.6) and is used because electrons passing through the Schottky barrier do so sufficiently quickly that they do not polarize the lattice [16, 7].

In this regime the current is

$$J = -q\mu N_{\mathrm{D}}E_0\exp\left(-\frac{q}{kT}(\phi_{\mathrm{b}} - \Delta\phi_{\mathrm{b}})\right). \qquad (16)$$

We note that this is the form of the Richardson–Schottky equation applicable in materials with very short mean free paths and has been previously derived by Simmons [3]. This form of the Richardson–Schottky equation has already been been noted as the appropriate one for

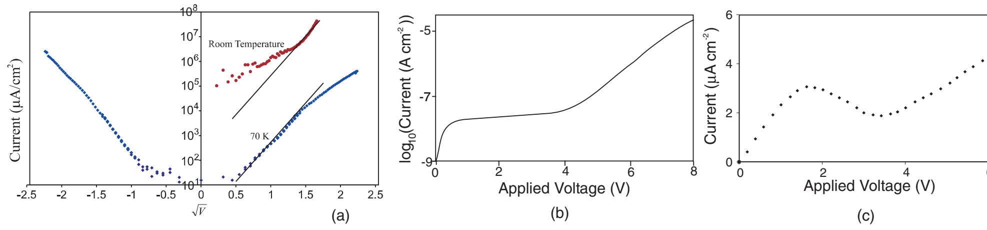


**Figure 2.** (a) Leakage current data from 70 nm thick Au–BST–$SrRuO_3$ film at room temperature and at $T = 70$ K; (b) Scott *et al* [10]: leakage current of BST thin film showing flat plateau followed by Schottky regime; (c) Watanabe *et al* [11]: leakage current of SBT film showing negative differential resistivity followed by Schottky regime.

(This figure is in colour only in the electronic version)

ferroelectric thin films and has been used successfully to describe leakage current in BST thin films by Zafar [8].

In a previous study on Au–BST–$SrRuO_3$ capacitors [9] we incorrectly assumed that the high-voltage regime corresponded to tunnelling through the space-charge region in the electrode because of the correspondence between the threshold for the high-voltage regime and the applied voltage at which the potential drop across the electrode equalled the barrier height; i.e., it was noted that

$$V_{\mathrm{T}} = \frac{\phi_{\mathrm{b}}}{2\delta_{\mathrm{eff}}}\left(2\delta_{\mathrm{eff}} + \frac{d}{\varepsilon_{\mathrm{s}}\varepsilon_0}\right). \tag{17}$$

We note however that the present model predicts the same thickness dependence of the threshold voltage: $V \propto (2\delta_{\mathrm{eff}} + \frac{d}{\varepsilon_{\mathrm{s}}\varepsilon_0})$. Low-temperature measurements on the 70 nm film from this study show that the high-voltage regime is Schottky-like both at room temperature and at 70 K (figure 2(a)).

The low-field expression derived above (equations (15) and (16)) can account for two kinds of low-field behaviour observed in ferroelectric thin films: either a sharp increase followed by a heretofore unexplained flat plateau (which is the behaviour of the equation when the donor concentration is very high) or a negative differential resistivity (observed for lower donor concentrations).

An example of the first kind of behaviour (figure 2(b)) is found in the data of Scott *et al* [10] on BST thin films, whereas a good example of the second (figure 2(c)) can be found in the SBT samples of Watanabe *et al* [11] or in PZT in the data of Scott *et al* [12] or Chen *et al* [13]. Previous attempts to explain these effects have invoked the filling and emptying of trap states. In both figures 2(b) and (c) it can be seen that the voltage threshold for crossover from the low-voltage regime to the high-voltage regime is around 4 V. From equation (14), if the film is 200 nm thick and has a dielectric constant of 1000 and a 10 nm depletion width, a 4 V threshold implies an oxygen vacancy concentration of around $10^{20}$ cm$^{-3}$, in good agreement with typical values for ferroelectric thin films which vary from $10^{18}$ to $10^{21}$ cm$^{-3}$ [14, 1]. We note that although we have assumed a uniform oxygen vacancy distribution this is not the case for most ferroelectric thin films which in reality have considerably more oxygen vacancies near the surface than in the bulk of the film. The mobility $\mu$ shows considerable variation from sample to sample, from $SrTiO_3$ single crystals with electron mobilities of 0.1 cm$^2$ V$^{-1}$ s$^{-1}$ to BST thin films with mobilities of 0.001 cm$^2$ V$^{-1}$ s$^{-1}$ [8]. For BST–Pt junctions the barrier height had been estimated at around 0.6–0.7 eV ($0.7 \pm 0.2$ [15], $0.65 \pm 0.06$ [8]). Using in equation (13) a mobility of 0.1 cm$^2$ V$^{-1}$ s$^{-1}$, barrier height of 0.65 eV, depletion width of

10 nm, oxygen vacancy concentration of $10^{20}$ cm$^{-3}$ and dielectric constant of 1000 we find a threshold current at room temperature of approximately $3 \times 10^{-8}$ A cm$^{-2}$, in good agreement with that found experimentally in figure 2(b) [10]. In reality variations in all these parameters from one sample to another are considerable, as are the observed current magnitudes.

On taking the derivative with respect to voltage of our low-field expression, we find that

$$\frac{\partial J}{\partial V} = \frac{q\mu N_\mathrm{D}}{2\delta_\mathrm{eff}\varepsilon_\mathrm{s}\varepsilon_0 + d}\left(1 - (E_0)^2\frac{\varepsilon_\mathrm{s}\varepsilon_0}{kTN_\mathrm{D}}\right)\exp\left(-\frac{q}{kT}\left(\phi_\mathrm{b} - \frac{qN_\mathrm{D}w^2}{2\varepsilon_\mathrm{s}\varepsilon_0} + \frac{\varepsilon_\mathrm{s}\varepsilon_0}{2qN_\mathrm{D}}(E_0)^2\right)\right).$$

The exponential term decreases with voltage towards the value $\exp(-\frac{\phi_\mathrm{b}}{kT})$ but is always positive. If the other voltage-dependent term remains positive, the low-voltage leakage current characteristic will be a rapid increase which plateaus off into an ohmic regime until such voltage that the barrier begins to be lowered by the Schottky effect. However, if the voltage is greater than

$$V > (2\delta_\mathrm{eff}\varepsilon_\mathrm{s}\varepsilon_0 + d)\sqrt{kT\varepsilon_\mathrm{s}^{-1}\varepsilon_0^{-1}N_\mathrm{D}} \tag{18}$$

the differential resistivity will become negative. We should not forget, however, the condition that this conduction mechanism only operates when

$$V < 2qN_\mathrm{D}w_2\left(2\delta_\mathrm{eff} + \frac{d}{\varepsilon_\mathrm{s}\varepsilon_0}\right) \tag{19}$$

and so negative differential resistivity is observed only if

$$\sqrt{\frac{\varepsilon_\mathrm{s}\varepsilon_0 kT}{4q^2N_\mathrm{D}w_2^2}} < 1. \tag{20}$$

If the film is 200 nm thick and has a dielectric constant of 1000 and a 10 nm depletion width, at room temperature this condition implies that negative differential resistivity will only be observed for oxygen concentrations greater than $3 \times 10^{18}$ cm$^{-3}$, though it is also noticeable that the magnitude of the effect will be quite small for high vacancy concentrations. Typical values for oxygen vacancy concentrations in titanate thin films are between $10^{18}$ and $10^{21}$ cm$^{-3}$ [14, 1], so in principle most ferroelectric films might be expected to display negative differential resistivity to some extent, though in highly oxygen deficient films the effect may be too small to be noticed.

Within the Schottky injection regime one of the most interesting things to note is the role that the dielectric constant plays in determining the temperature dependence of the current. We can show that when the sample is below a ferroelectric phase transition and the dielectric constant is increasing rapidly with temperature a regime of positive temperature coefficient of resistivity (PTCR) may be observed. For this purpose we simplify the Schottky regime current expression by taking

$$\Delta\phi_\mathrm{b} = \sqrt{\frac{q}{4\pi\varepsilon_\infty\varepsilon_0}(-E_0)} \tag{21}$$

and taking the derivative with respect to temperature we find that

$$\frac{\partial J}{\partial T} = J\left(-\frac{q}{kT}\left[\frac{1}{T}(\phi_\mathrm{b} - \Delta\phi_\mathrm{b}) - \Delta\phi_\mathrm{b}\left(\frac{\partial\varepsilon_\mathrm{s}}{\partial T}\left(\frac{\delta_\mathrm{eff}}{2\delta_\mathrm{eff}\varepsilon_\mathrm{s}\varepsilon_0 + d}\right)\right)\right]\right). \tag{22}$$

The term $\frac{1}{T}(\phi_\mathrm{b} - \Delta\phi_\mathrm{b})$ is always positive. For a PTCR effect to be observed above a certain applied voltage $\frac{\partial\varepsilon_\mathrm{s}\varepsilon_0}{\partial T}(\frac{\delta_\mathrm{eff}}{2\delta_\mathrm{eff}\varepsilon_\mathrm{s}\varepsilon_0 + d})$ must be large and positive, and hence the effect should be seen only below $T_C$. Whether or not PTCR behaviour is observed depends on the barrier height and the screening length in the metal, with PTCR behaviour more likely to occur for

capacitors with lower barrier heights and longer screening lengths. The PTCR effect predicted here does not involve grain boundaries, as distinct from the PTCR effect in $BaTiO_3$ ceramics (for a review see Huybrechts *et al* [18]) which as shown by Sinclair and West [17] has both bulk and grain boundary contributions. Hwang [19] has observed PTCR effects in films with Ir electrodes, whereas similar films with Pt electrodes did not display PTCR behaviour. This is consistent with our model because Ir has a significantly lower workfunction than Pt (4.23 eV compared to 5.3 eV [20]).

Thus with a single model we have explained many unusual leakage current features in ferroelectric capacitors. Further development of this model will require detailed fitting of leakage current in samples in which the parameters of the model are independently measured using other experimental techniques, for example by combining optical and electrical measurement techniques to better determine the nature of the oxygen vacancy trap levels.

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