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Electric-pulse-induced reversible resistance in doped zinc oxide thin films

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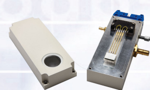

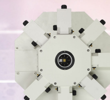

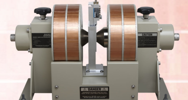

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Electric-pulse-induced reversible resistance in doped zinc oxide thin films

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Nonvolatile, electric-pulse-induced resistance switching is reported on S and Co doped ZnO thin films deposited on different substrates using magnetron sputtering and laser ablation. Two resistance states were obtained by applying voltage pulses of different polarity. The switching was observed regardless of the substrate, dopant species, or microstructure of the samples. In the Co doped ZnO samples, the two resistance states are remarkably stable and uniform. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2437688]

Resistance switching and voltage controlled negative resistance phenomena have been observed since the 1960s on materials such as amorphous semiconductors[1–3] and ZnSe–Ge heterostructures,[4] as well as on a variety of oxides such as Nb_2O_5,[5] Al_2O_3,[6] Ta_2O_5,[7] TiO_2,[8] and NiO.[9,10] An apparently related phenomenon, known as electric-pulse-induced resistance switching (EPIR), has been recently reported in $Pr_{0.7}Ca_{0.3}MnO_3$ thin films at room temperature.[11] In this effect, a nonvolatile change of the electrical resistance between two electrodes is observed after applying an electric pulse. Thus, the EPIR effect is different from "conventional" resistance switching, where the effect vanishes as the bias is reduced.[12] Furthermore, the direction of the resistance change (i.e., either increased or decreased resistance) depends on the polarity of the electric pulse in EPIR.

In this work, we report a reproducible EPIR effect in doped ZnO. Thin films of ZnO doped with sulfur and cobalt and deposited using magnetron sputtering and laser ablation, respectively, exhibit the switching behavior characteristic of the EPIR effect, without the need of previous sample training. In particular, in the Co doped ZnO samples, the resistance is remarkably stable and uniform in both the low and the high resistance states.

Thin films of ZnO were fabricated by two methods: (a) pure ZnO and sulfur doped ZnO films (S:ZnO) were deposited by radio frequency magnetron sputtering at room temperature in argon atmosphere on SiO_2/Si and on Si substrates and (b) cobalt doped ZnO films (Co:ZnO) were grown on Si_3N_4/Si substrates by pulsed laser deposition (PLD) at 400 °C under 5 Pa of nitrogen (using a ZnO target with a nominal Co content of 15%). The thickness of all the films was about 100 nm, as estimated form secondary ion mass spectrometry and Rutherford backscattering spectrometry measurements. X-ray diffraction (XRD) patterns corresponded to the ZnO wurtzite structure for all the films. The sputtered ZnO and S:ZnO films were polycrystalline although textured with the *c* axis ($c=0.507\pm0.001$ nm) perpendicular to the substrate. The Co:ZnO films were found to be highly oriented, with the *c* axis ($c=0.516\pm0.001$ nm) aligned perpendicular to the substrate, and exhibit only (002) and (004) XRD lines. For the electrical transport measurements, leads were soldered with Ag epoxy to round Au contacts sputtered on the top surface of the samples. A two-probe configuration, in constant voltage mode, was employed. Figure 1 illustrates schematically the setup used for *I-V* characterization and pulse application. The current-voltage curves (*I-V* curves) were constructed using tension pulses of increasing amplitude and about 1 s duration, each followed by about half a second without applied tension in order to minimize potential Joule heating. To study the EPIR effect, short high-voltage pulses (40 ms) were applied using a Keithley 220 programmable current source, and current flow upon low voltage bias ($\leqslant 1$ V) was monitored through the voltage drop across a known resistor (1 kΩ) using a Keithley 182 nanovoltmeter. Positive bias was defined by the current flowing from electrode A to electrode B (see Fig. 1), and negative bias was defined by the opposite direction. The measurements were made in a vacuum of ~1 mTorr and at room temperature.

Figure 2(a) shows the *I-V* curve for a S:ZnO film deposited by sputtering on a SiO_2/Si substrate. The resistance as a function of applied voltage, calculated from the *I-V* data, is shown in the inset. Initially, the film exhibited high resistivity. However, the number of charge carriers could be en-

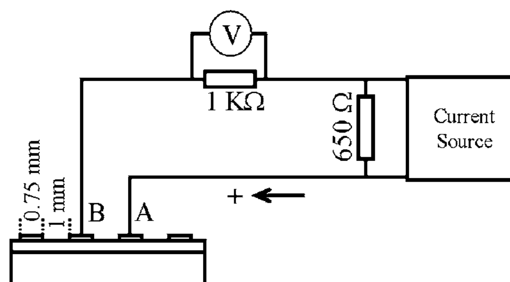

FIG. 1. Setup used for pulse application and current-voltage (*I-V*) characterization.

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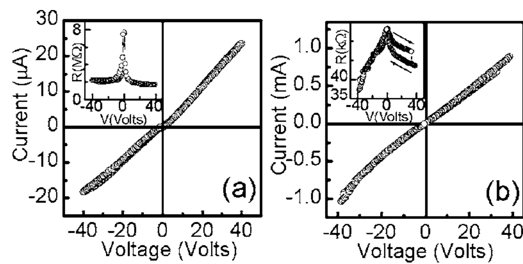

FIG. 2. Current voltage (*I*-*V*) characteristics of (a) a sulfur doped ZnO sample deposited on SiO_2/Si substrate (measured under UV illumination) and (b) a cobalt doped ZnO film deposited on Si_3N_4/Si. The inset shows the calculated resistance as a function of voltage.

hanced, and the resistivity reduced, by irradiation before (for ~1 h) and during measurement with ultraviolet (UV) light. For this purpose, an OSRAM™ ULTRAVITALUX lamp of 300 W, with two main intensity peaks at 310 and 360 nm, was used. An undoped ZnO sample deposited on a SiO_2/Si substrate showed a similar *I*-*V* curve (not shown), but with much higher values of resistance (between 300 and 500 MΩ). The *I*-*V* curve (measured in the dark) of the S:ZnO film deposited on a Si substrate (not shown) exhibited stronger nonlinearity and lower resistance (~2– ~100 KΩ, depending on applied voltage) than the sample shown in Fig. 2(a). Figure 2(b) shows the *I*-*V* curve for a Co:ZnO film deposited by PLD on a Si_3N_4/Si substrate. As can be seen from the inset in Fig. 2(b), there is a resistance switch at the positive branch of the *I*-*V* curve. This feature is not detected in the *I*-*V* curves of the films deposited by sputtering.

Figures 3(a) and 3(b) show the voltage pulse driven high-resistance–low-resistance switching for the S:ZnO samples deposited on SiO_2/Si and Si, respectively. The measurement process consisted in using a "read" voltage of +0.1 or +1 V to monitor the resistance of the sample and a sequence of 15 consecutive pulses of ±40 V and 40 ms of duration each to switch the resistance. The number of pulses in the sequence was chosen in order to observe a clear and reproducible effect, although a series of less pulses would also induce the resistance switch. Figure 3(a) shows that, after the group of 15 pulses of +40 V is applied, the resistance switches to a low resistance state. Then, after the series of 15 pulses of −40 V, the resistance reverts to its original higher value. Both films switched to low resistance after (+) pulses and reverted to high resistance after (−) pulses. Figure 3(a) also shows that the high resistance values are unsteady, showing a capacitive effect common in resistive materials. Figure 3(b) shows a similar effect, but without the capacitive decay, in the more conductive ZnO/Si sample. The undoped ZnO sample analyzed in the present study did not exhibit the switching effect. Figure 3(c) shows the voltage pulse driven, high-resistance–low-resistance switching for the Co:ZnO sample. It is observed that the film switches to a low resistance state after (+) pulses and reverts to a high resistance state after (−) pulses in a similar way as the S:ZnO samples. However, in the case of the Co:ZnO sample, the resistance is remarkably more stable and uniform in both the low and the high resistance states.

We do not have yet a definitive explanation for the EPIR effect in doped ZnO. Indeed, the physical origin of the resistance switch in other materials, where the effect was observed previously, is still under intense debate and different

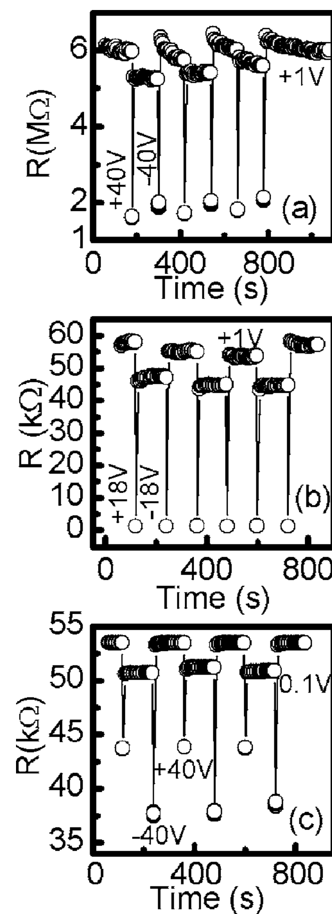

FIG. 3. Resistance response of ZnO films in (a) sulfur doped ZnO film on SiO_2/Si substrate (measured under UV illumination at 300 K) when an alternate series of voltage pulses of +40 and −40 V is applied every 120 s, (b) sulfur doped ZnO film on Si substrate (measured in the dark at 290 K) when an alternate series of voltage pulses of +18 and −18 V is applied every 120 s, and (c) cobalt doped ZnO film deposited by pulsed laser deposition on Si_3N_4/Si (measured in the dark at 290 K) when an alternate series of voltage pulses of +40 and −40 V is applied every 120 s.

models have been proposed.[11,13–17] In previous works on the electrical transport properties of polycrystalline ZnO thin films,[18–20] we have shown that the electrical conductivity of ZnO is modulated by effects induced by bulk and grain boundary defect states. In order to guide future work, we have adopted the working hypothesis that defect states (in the bulk and at the ZnO/metal interfaces) are responsible, directly or indirectly, for the resistance switching effect. Traps sufficiently deep to have trapping times of tens of seconds or more have been detected in ZnO,[21,22] and carrier trapping has been recently suggested as the origin of resistance switching in manganites.[23] Further work is in progress to test the validity of these assumptions and elucidate the physical mechanisms responsible for resistance switching in polycrystalline ZnO thin films.

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