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Epitaxial diodes of a half-metallic ferromagnet on an oxide semiconductor

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We report on the fabrication and electrical characterization of epitaxial Schottky diodes of a half-metallic ferromagnet on an oxide semiconductor. $La_{0.67}Sr_{0.33}MnO_3$ thin films are grown by pulsed laser deposition on niobium-doped $SrTiO_3$ semiconductor substrates with two doping concentrations and a TiO_2 surface termination. The current across the diodes is dominated by thermionic emission and shows high rectification and low reverse bias leakage. At room temperature, the Schottky barrier height is 0.95 eV (0.65 eV) and the ideality factor is 1.08 (1.18) for the diodes with a low (high) doped semiconductor. With decreasing temperature the Schottky barrier height decreases and the ideality factor increases. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1669255]

Half-metallic ferromagnets like $La_{0.67}Sr_{0.33}MnO_3$ (LSMO)[1] are promising materials for spintronic devices such as magnetic tunnel junctions.[2,3] State-of-the-art tunnel junctions consisting of LSMO and $SrTiO_3$ barriers (STO) have shown tunnel magnetoresistance of 1800% at 4.2 K.[4] Unfortunately, the magnetoresistance vanishes at 280 K,[4] despite the fact that the Curie temperature is 360 K.[5] This is believed to be due to a reduced high temperature spin polarization at the interface.[4,6]

Tunneling has also been exploited to achieve successful spin injection into a semiconductor. Injection is either by tunneling through a reverse biased Schottky diode contact of a conventional ferromagnetic metal (Fe) on an *n*-type semiconductor,[7] or tunneling through a thin (Al_2O_3) tunnel barrier inserted between the ferromagnet and the semiconductor.[8] Half-metallic ferromagnets are also interesting candidates for spin injection into a semiconductor. However, if transport is by interface sensitive tunneling, we may expect a degradation of the spin injection at higher temperature, much in the same way as the decay of tunnel magnetoresistance in LSMO-based magnetic tunnel junctions.[4] However, it was suggested[9] that direct spin injection via diffusive transport might be possible when ferromagnetic contacts with near 100% spin polarization are used. It is therefore highly relevant to study transport across interfaces between a half-metallic ferromagnet and a semiconductor and establish the transport mechanism.

The Schottky barrier that forms in a metal–semiconductor contact is a critical part of spintronic devices such as the spin-valve transistor and the magnetic tunnel transistor.[10] These devices have shown large magnetotransport effects, even at room temperature, due to highly spin-dependent transmission of hot electrons through the metallic base of the transistor. Since it was shown [11] that interfaces contribute little to the spin dependence of hot-electron transmission, we anticipate that transistors using a half-metallic metal base should also operate at room temperature. A first step towards realization of such a device is the fabrication of a Schottky diode of a half-metallic ferromagnet on a semiconductor. If the structure can be grown with high structural quality, the scattering by defects is reduced and the output current of the transistor can be increased.

Here we report on the fabrication and electrical characterization of epitaxial diodes of half-metallic LSMO on a lattice matched Nb-doped $SrTiO_3$ (STO:Nb), which is an oxide semiconductor. While insulating STO is commonly used as a substrate for the growth of epitaxial LSMO films, the use of a Nb-doped STO semiconductor to form diodes with a half-metallic ferromagnet has hitherto not been reported. Shimizu and Okushi[12] have reported characteristics of high-quality Schottky diodes using Nb-doped $SrTiO_3$ semiconductor substrates, but only in combination with regular metals like Cu and Au.[12]

The diodes were grown on [001] oriented Verneuil-type STO:Nb single crystal substrates. The niobium doping density used here is 0.1 wt % or 0.01 wt %, corresponding to $SrTi_{(1-x)}Nb_xO_3$ with $x=0.002$ and 0.0002, respectively. These *n*-type semiconductor substrates were treated to achieve a TiO_2 termination.[13] An ultrasonic bath of deionized water (30 min) hydrates any SrO present at the STO surface. The hydrate is dissolved in buffered HF leaving a TiO_2 terminated surface. Atomic force microscopy (AFM) shows only steps of one unit cell height, confirming the single termination. The low doped substrates were annealed at 950 °C in 1 bar of oxygen to decrease the step-edge density. The high doped substrates were not annealed, because AFM indicated that it causes the niobium to diffuse to the surface.

The LSMO films are grown on the TiO_2 terminated STO:Nb substrates by pulsed laser deposition using a KrF excimer laser (248 nm) with an energy density of 1 J/cm^2 at a repetition rate of 3 Hz. The distance between substrate and target is 4 cm. The substrate temperature is kept at 750 °C during deposition, while controlling the oxygen pressure to 0.3 mbar. After deposition the oxygen pressure is raised to 1 bar and the substrate is cooled to room temperature at a rate

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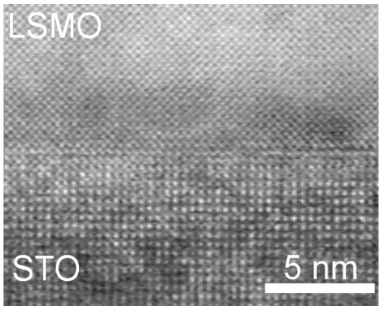

FIG. 1. Cross sectional transmission electron microscopy image of a LSMO film on STO.

of 10 °C/min. The LSMO films are patterned into 350 $\times 700\ \mu\mathrm{m}^2$ rectangles by standard photolithography and ion beam etching. Electrical contacts to the LSMO are made by ultrasonic Au wire bonding. Ohmic contacts to the substrate are obtained by ultrasonic Al wire bonding. The Al bonds were also used for four-terminal resistivity measurement of the substrates.

Transmission electron microscopy (Fig. 1) shows that the LSMO has a sharp interface with the STO substrate and is epitaxial. This is confirmed by x-ray diffraction measurements that show only [001] and higher order reflections of the LSMO and the substrate. The LSMO films are thus epitaxial and [001] oriented.

The resistivity of the STO:Nb semiconductor substrates at 300 K was measured to be 1.6 Ω cm for the low doped substrates and 0.038 Ω cm for the high doping. The resistivity is reduced with decreasing temperature following a $T^{2.8}$ dependence. This is in agreement with a temperature independent carrier concentration, and a carrier mobility with $T^{-2.8}$ dependence, as found in Hall measurements.[12,14]

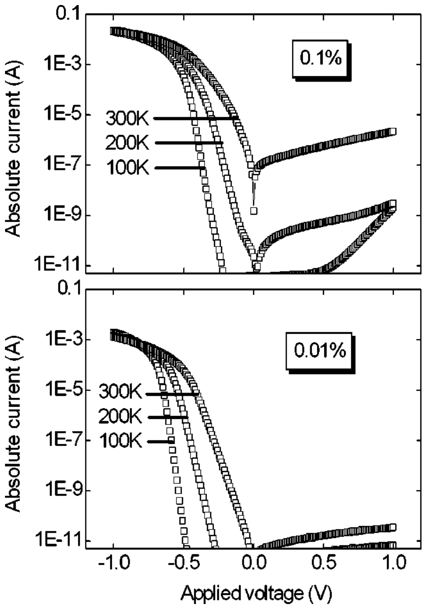

FIG. 2. Current–voltage characteristics of LSMO/STO:Nb Schottky diodes at 100, 200, and 300 K for niobium doping concentrations of 0.1 wt % (top panel) and 0.01 wt % (bottom panel). The absolute value of the current is plotted and $V>0$ corresponds to positive voltage on the semiconductor.

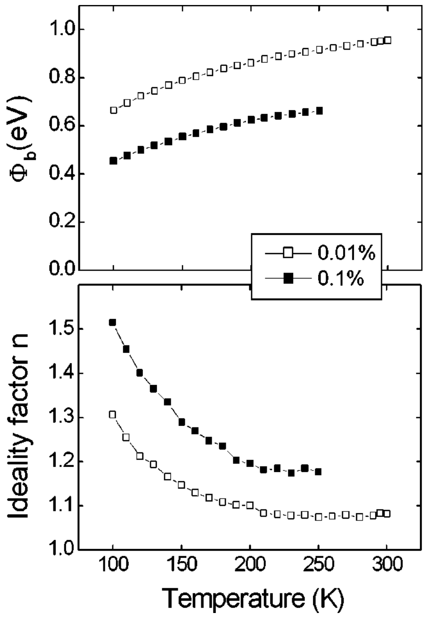

FIG. 3. Temperature dependence of the Schottky barrier height Φ_b (top panel) and the ideality factor n (bottom panel) of LSMO/STO:Nb Schottky diodes. Open symbols represent diodes with 0.01 wt % Nb-doped semiconductor, the solid symbols are for STO:Nb with 0.1 wt % Nb doping.

The current–voltage characteristics of the diodes (Fig. 2) are highly rectifying. For diodes with a low doped semiconductor (bottom panel), the reverse bias current at +1 V is on the order of 10 pA, and the forward bias current is more than 7 orders of magnitude larger. The direction of the rectification is in agreement with an *n*-type semiconductor Schottky diode. For diodes with high doped STO:Nb (top panel) the reverse bias current at +1 V is around 1 μA at room temperature and the rectification is smaller. However, the reverse bias current drops fast with temperature and decreases to a few nA at 200 K.

The Schottky barrier height (Φ_b) and ideality factor (n) can be extracted by fitting the data to the well-known expression for the thermionic emission current density[15,16]

$$J=A^*T^2\exp\left(-\frac{q\phi_b}{kT}\right)\left[\exp\left(\frac{qV}{nkT}\right)-1\right], \tag{1}$$

in which A^* is the Richardson constant with a value of 156 $\mathrm{A\,cm^{-2}\,K^{-2}}$ for STO:Nb.[12] The dependence of the Schottky barrier height and the ideality factor on temperature is shown in Fig. 3 (see also the note in Ref. 17). The ideality factor of the diodes with low doped semiconductor is 1.08 at room temperature, implying nearly perfect thermionic emission. For the high doped semiconductor the ideality factor of 1.18 is somewhat larger. The Schottky barrier heights at room temperature are 0.95 and 0.65 eV, respectively, for low and high doped STO:Nb. With decreasing temperature the Schottky barrier height decreases and the ideality factor increases. Such a temperature dependence and an ideality factor that deviates from unity was also reported for Au/STO:Nb diodes.[12] It was shown to be due to the presence of an intrinsic low permittivity layer at the STO surface, and the increase of the dielectric constant of STO at low temperature.

Since we observe similar behavior for the LSMO/Nb:STO diodes, the same explanation may also apply here.

In conclusion, we have fabricated high-quality epitaxial Schottky diodes of a half-metallic ferromagnet (LSMO) on semiconductor STO:Nb substrates of different doping density. Electrical transport across the interface is dominated by thermionic emission and the diodes are highly rectifying and have low reverse bias current. These diodes are useful building blocks for spintronic devices, and are extremely suitable for use as collector Schottky barrier in a magnetic tunnel transistor.

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