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Magnetoresistance switch effect in a multiferroic $Fe_3O_4/BaTiO_3$ bilayer

M. Ziese[a]
Division of Superconductivity and Magnetism, University of Leipzig, 04103 Leipzig, Germany

A. Bollero
SPINTEC, CEA/CNRS, 38054 Grenoble Cedex 9, France

I. Panagiotopoulos
Department of Materials Science and Engineering, University of Ioannina, Ioannina 45110, Greece

N. Moutis
Institute of Materials Science, NCSR "Demokritos," Agia Paraskevi Attiki, 15310, Greece

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Multiferroic bilayers composed of a magnetite (Fe_3O_4) and a $BaTiO_3$ layer show nonlinear current-voltage characteristics in current perpendicular to plane configuration. The magnetoresistance of the bilayers is strongly bias dependent and can be switched from negative at low bias to positive at large bias. It is shown that these effects do not arise from charge-carrier modulation in the magnetite layer by an electric field effect. Therefore both the nonlinear transport characteristics and the switchable magnetoresistance are attributed to interfacial transport phenomena. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2206121]

Oxide spintronics has attracted an enormous attention from researchers worldwide, since various unforeseen magnetoresistance effects were discovered[1,2] and since oxides hold the potential of combining materials with different characteristics such as ferromagnets, ferroelectrics, and superconductors in a single device using heteroepitaxial growth techniques. In such devices the interface will play a decisive role in determining the functionality of the heterostructure and accordingly in recent years various studies of the transport properties at oxide interfaces have emerged, see, e.g., Refs. 3–10. The ferrimagnet magnetite (Fe_3O_4) has a high spin polarization[9,11,12] and a high Curie temperature of 860 K and is a potential candidate for applications in spintronics devices. In the present paper magnetotransport properties of the magnetite/$BaTiO_3$ interface are reported. A bias dependent switchable magnetoresistance was found.

In this work two types of multilayer structures were investigated. First, so-called "direct" structure consisted of a magnetite film deposited on a $MgAl_2O_4$ (001) substrate and covered with a $BaTiO_3$ layer and a gold capping layer, see inset of Fig. 4. Second, so-called "inverse" structure was fabricated by depositing a $BaTiO_3$ and then a magnetite layer on a conducting Nb(0.1%):$SrTiO_3$ (001) substrate, see inset of Fig. 1; in a first deposition run a gold layer was deposited on the substrate's unpolished backside. Layer thicknesses were 50 nm for magnetite, 300 nm for $BaTiO_3$, and 100–150 nm for gold. The films were fabricated by pulsed laser deposition from stoichiometric polycrystalline targets and a gold target, respectively. Oxygen partial pressure was 9×10^{-6} mbar for the oxide deposition. In the case of the direct structure both magnetite and $BaTiO_3$ layers were deposited at a substrate temperature of 450 °C, whereas in the case of the inverse structure the $BaTiO_3$ layer was grown at 750 °C. The gold layers were deposited at 450 °C at a pressure of 5×10^{-7} mbar. The growth of the Fe_3O_4 and $BaTiO_3$ layers was monitored by *in situ* reflection high energy electron diffraction (RHEED) measurements. The magnetite layers as well as the $BaTiO_3$ layers grown at 750 °C were epitaxial; on the contrary, the $BaTiO_3$ layers grown at 450 °C were amorphous. The $BaTiO_3$ layers were transparent with an in-plane resistance of about 250 G Ω (corresponding to a resistivity of about 7.5×10^6 Ω cm) in the case of the layers fabricated at 750 °C and $>10^4$ G Ω for the layers fabricated at 450 °C. The samples were patterned by conventional photolithography and ion-beam etching to define bridges of width 50 μm in standard four-point configuration for electrical characterization, see the schematic layout in the inset of Fig. 1. Electrical contacts were made with silver paint on the magnetite layer for the current and voltage contacts and with silver paint on the gold layers for the gate and backside contacts. One unpatterned inverse structure was measured and found to show qualitatively the same results as the patterned sample. All transport measurements were performed with a four-point technique in a continuous flow cryostat equipped with a 9 T superconducting solenoid. The magnetic field was applied parallel to the magnetite layer. From magnetization measurements the Verwey temperature T_V was determined to be 115 K.

Current (I)-voltage (U)-characteristics of an inverse $Fe_3O_4/BaTiO_3$ structure are shown for forward bias in Fig. 1(a). Forward bias is defined with the positive voltage applied to the conducting Nb:$SrTiO_3$ substrate. The *I-U* curves are nonlinear with the nonlinearity becoming especially pronounced below 120 K. Furthermore the *I-U* curves have a strong asymmetry, as seen in Fig. 2(a). At low bias the *I-U* curves are linear and an Ohmic resistance R was defined. This is shown in Fig. 1(b) as a function of temperature; a clear change of slope is observed in the $R(T)$ curve at about 120 K that is attributed to the Verwey temperature. These data show that the transport properties of the inverse heterostructure are dominated by the interface between the $BaTiO_3$ and magnetite layers, since the bulk transport properties are linear and since the signature of the Verwey transition appears in the *I-U* curves. The data are similar to those on

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[a] Electronic mail: ziese@physik.uni-leipzig.de

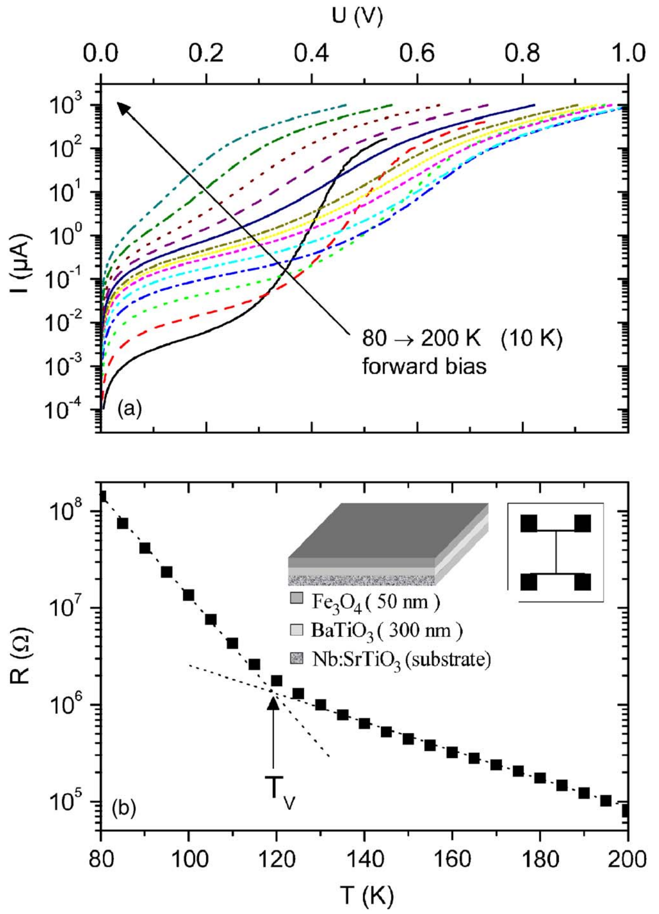

FIG. 1. (Color online) (a) Forward bias current (*I*)-voltage (*U*) curves of an inverse Fe_3O_4–$BaTiO_3$ bilayer recorded in zero field; the temperature ranges from 80 to 200 K in steps of 10 K. (b) Resistance of the structure as a function of temperature. The insets show the multilayer structure and the layout of the patterned device, respectively.

magnetite/Nb:$SrTiO_3$ interfaces[9] with the difference that in the present case the nonlinearity of the current-voltage curves is much stronger.

The application of a magnetic field shifts the *I*-*U* curves as indicated in Fig. 2. The magnetocurrent ratio $I(B)/I(0)$ for applied fields up to 8 T is bias dependent as shown in Figs. 2(b) and 2(c) for 90 and 130 K, respectively. Main feature of the data is a crossover of the magnetoconductance from positive values at low bias voltage to negative values at high bias voltage.

Figure 3 shows the conventional magnetoresistance ratio defined as $[U(B)-U(0)]/U(0)$ measured at temperatures of 90 and 130 K at constant forward and reverse bias currents. The magnetoresistance has a strong bias dependence which proves that it is an inherent property of the barrier and is not due to bulk scattering processes in the magnetite film. These curves clearly show that the sign of the magnetoresistance can be switched from negative to positive by applying low and high bias currents, respectively.

The positive magnetoconductance at low bias voltages is similar to that observed for magnetite/Nb:$SrTiO_3$ interfaces.[9] In that case the magnetoconductance was attributed to the spin dependence of the Schottky-barrier height at the Fe_3O_4/Nb:$SrTiO_3$ interface that—within the two-current model—leads to a linear field dependence of the current density with a prefactor proportional to the spin polarization.[9] Applied to the $Fe_3O_4/BaTiO_3$ interface considered here this model yields spin-polarization values of $P=1\pm0.5$.

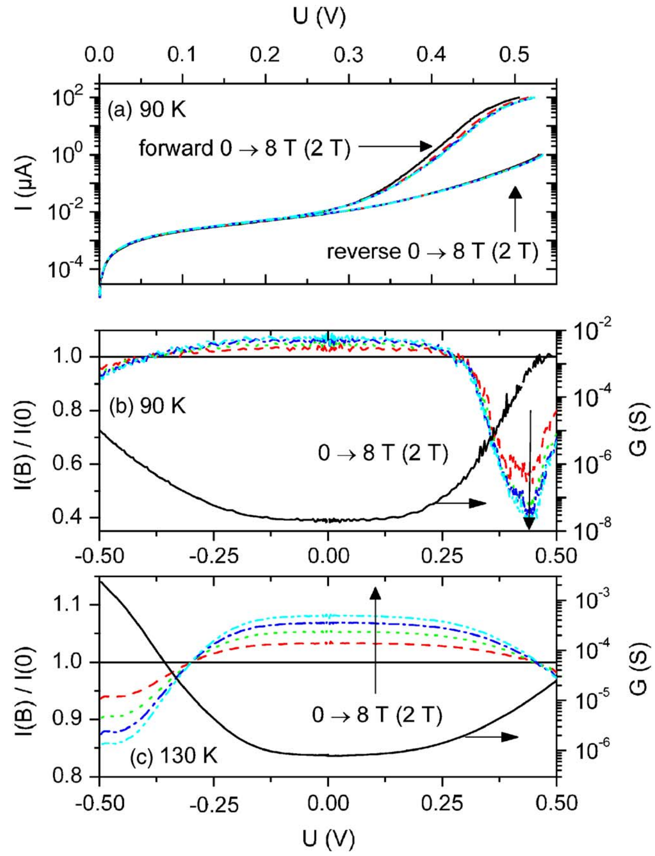

FIG. 2. (Color online) (a) Forward and reverse bias *I*-*U* curves at 90 K for magnetic fields of 0, 2, 4, 6, and 8 T. [(b) and (c)] Magnetocurrent ratio $I(B)/I(0)$ (left axis) as a function of bias voltage at 90 and 130 K for magnetic fields of 0, 2, 4, 6, and 8 T. For comparison the differential conductance $G=dI/dV$ in zero magnetic field is shown (right axis).

The magnetotransport behavior at higher current bias, especially the sign change of the magnetoresistance, might be related to electric field effects in the magnetite layer, i.e., changes of the carrier density due to the electric field in the adjacent $BaTiO_3$ layer, to current redistribution processes,[13] or to current breakdown mechanisms at the interface.[14] In order to investigate the first mechanism the resistance modu-

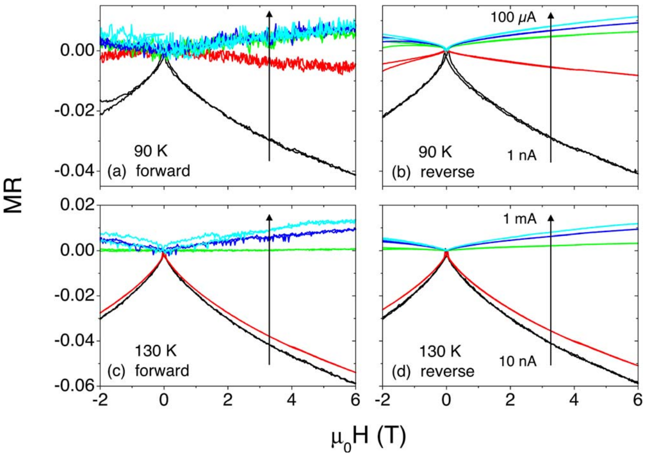

FIG. 3. (Color online) Magnetoresistance ratio $[U(B)-U(0)]/U(0)$ of an inverse Fe_3O_4–$BaTiO_3$ heterostructure measured at 90 and 130 K as a function of magnetic field at current bias values ranging from 1 nA(10 nA) to 100 μA(1 mA) in powers of 10.

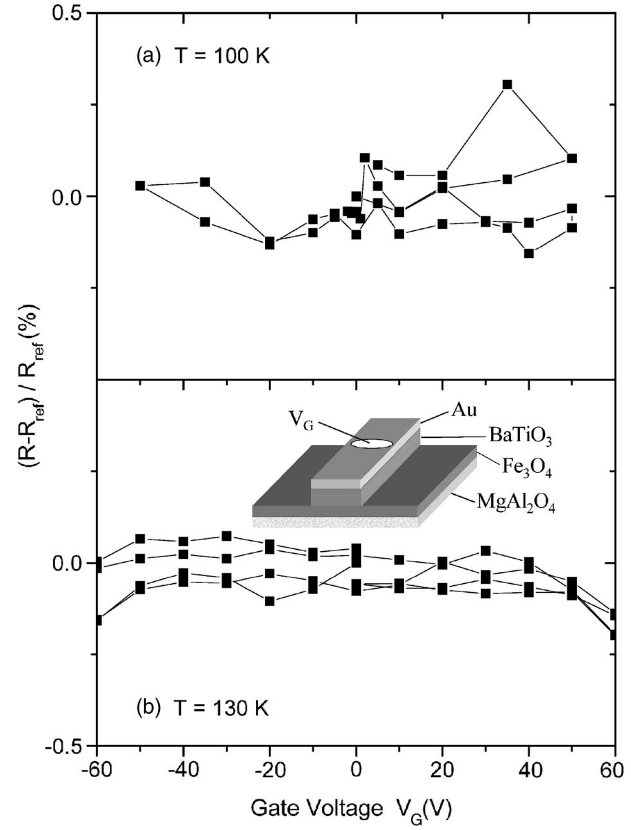

FIG. 4. Dependence of the resistance of the conducting magnetite channel in a direct structure on the gate voltage applied over the $BaTiO_3$ layer. The inset shows a schematic layout of the structure.

lation of the magnetite layer with applied bias voltage was measured in a direct structure. The resistance change versus gate voltage measured at 100 and 130 K is shown in Fig. 4. Within the experimental error the resistance of the magnetite layer does not depend on the applied gate voltage. This rules out the first explanation for the magnetoresistance sign change and shows that the electric field screening length in magnetite films is much smaller than 50 nm. In the second explanation it is considered that the current and voltage electrodes on the magnetite film are not at the same potential, since the resistance of the magnetite layer is rather high. The application of a magnetic field decreases the resistance of the magnetite layer and at the same time decreases the potential drop between the current and voltage electrodes. This is registered as a potential increase at the voltage contact due to the magnetic field, i.e., a positive magnetoresistance. This scenario is supported by the observation that the zero crossings of the current ratio for both bias directions as shown in Figs. 2(b) and 2(c) occur at similar values of the differential conductance *G*. The third explanation cannot be probed at the moment, since a model for the conduction processes in junctions at high bias voltages in the presence of a magnetic field has not yet been developed. However, we wish to point out that the magnetoresistance crossover is similar to findings for manganite/Nb:$SrTiO_3$ junctions.[15] It is known that Nb:$SrTiO_3$ forms an insulating surface layer under oxygen exposition at high temperature.[16] Therefore the magnetoresistance sign change might be an intrinsic feature of oxide structures with a ferroelectric or strongly dielectric interface barrier.

In summary, it has been shown that the magnetoresistance arising at the $Fe_3O_4/BaTiO_3$ barrier is strongly bias dependent; its sign can be switched from negative at low bias to positive at high bias. Future research should focus on a further understanding of the high bias regime.

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