ФІЗИКА І ХІМІЯ ТВЕРДОГО ТІЛА
Т. 8, № 3 (2007) С. 638-640

PHYSICS AND CHEMISTRY OF SOLID STATE
V. 8, № 3 (2007) P. 638-640

PACS: 68.55.-A, 75.50.GG, 73.40.CG, 73.40. LQ

ISSN 1729-4428

V. Lisauskas, B. Vengalis, K. Šliužienė, R. Butkutė

Nonlinear Electrical Properties of p-n Heterostructures Based on Ferrimagnetic Fe_3O_4

Semiconductor Physics Institute, A. Goštauto 11, LT-01108, Vilnius, Lithuania, E-mail: lisa@pfi.lt

We report preparation and investigation of p-n heterostructures consisting of ferrimagnetic p-type Fe_3O_4 layers grown on and n-type Si substrates and n-type layers of indium oxide (IO) and tin-doped indium oxide (ITO). The Fe_3O_4 films (d = 60 to 600 nm) were deposited in-situ by a reactive dc magnetron sputtering at T = 350÷450°C. Thin IO (ITO) films (d = 200÷500 nm) were prepared on YSZ(100) substrates at 250÷600°C by sputtering of pure In or In-Sn alloy (91:9) target. The Fe_3O_4/n-Si heterostructures demonstrated nonlinear current-voltage (I-V) dependencies in a wide temperature range (T = 78÷300 K) meanwhile similar I-V curves measured for the Fe_3O_4/(IO)ITO heterostructures showed asymmetry and nonlinearity only at T < 120K.

Key words: dc magnetron sputtering, Fe_3O_4 thin films, p-n heterostructures.

Стаття поступила до редакції 13.11.2006; прийнята до друку 15.06.2007.

Introduction

In recent years, an increasing research activity was focused on colossal magnetoresistance manganites ($La_{1-x}A_xMnO_3$, A = Ca, Sr, Ba), magnetite (Fe_3O_4), CrO_2 and other mixed valence ferromagnetic oxides. Presence of spin-polarized carriers is a unique property of all these oxides making them attractive for a number of applications in spin-electronic devices utilizing high spin polarization [1]. Spin-polarized electrons passing through intergrain boundaries, elaborated tunneling and Schottky barriers [2] as well as p-n heterostructures are expected to show a large value of magnetoresistance (MR).

Magnetite, Fe_3O_4, with an inverse cubic spinel structure (a = 0,8396 nm) exhibit electrical conductivity ($\rho \cong 10$ mΩcm at T = 300 K) due to a hopping of spin-polarized electrons between ferrimagnetically ordered Fe^{2+} and Fe^{3+} ion states. High Curie temperature ($T_c \cong 858$ K) of the oxide is an important advantage compared to other half-metallic oxides for room temperature applications. Fabrication of magnetic tunnel junctions using Fe_3O_4 thin films has attracted considerable attention over the past few years [3]. However, up to now there were only limited attempts to prepare magnetic p-n junctions containing Fe_3O_4. In this work, we report the results of preparation and investigation of p-n heterostructures consisting of p-type Fe_3O_4 layers grown on n-type conducting IO (ITO) layers, and n-type Si substrates.

I. Film Growth and Characterization

The p-n device structures were prepared by growing p-type Fe_3O_4 layers on both n-Si(111) substrates and n-type layers of either indium oxide (IO) or highly conducting tin-doped indium oxide (ITO). Fe_3O_4 thin films with typical thickness ranging from 60 to 600 nm were deposited in-situ by a reactive dc magnetron sputtering at T = 350÷450°C. A disk of metallic Fe (35 mm in diameter and 0,5 mm thick) was used as a target. Film growth was performed in Ar : O_2 gas mixture (10:1) ambient keeping partial oxygen pressure in the vacuum chamber of about 0,16 Pa. After deposition, the films were cooled down slowly to a room temperature under the same oxygen pressure conditions. To prevent possible film bombardment by high energy ions during deposition, the substrates were positioned in the "off-axis" configuration at a distance of 30-60 mm from the symmetry axis of the discharge and 20-25 mm above the target plane. It is important to note that in such a case, both thickness of the grown films and film composition (Fe and oxygen ratio) depended on the distance between the target and a certain position on the substrate.

Non-doped IO and tin-doped indium oxide (ITO) thin films with thickness ranging from about 200 nm to 500 nm were prepared on lattice-matched YSZ(100) substrates ($a_{IO} \cong a_{ITO}$ = (1,01–1,03 mm) $\cong 2a_{YSZ}$) by sputtering pure In or In-Sn (91:9) alloy targets. Temperature of the substrates during film growth was kept at 250÷600°C. Sputtering was performed in Ar:O_2 gas mixtures (4:1) at a pressure of about 5 Pa.

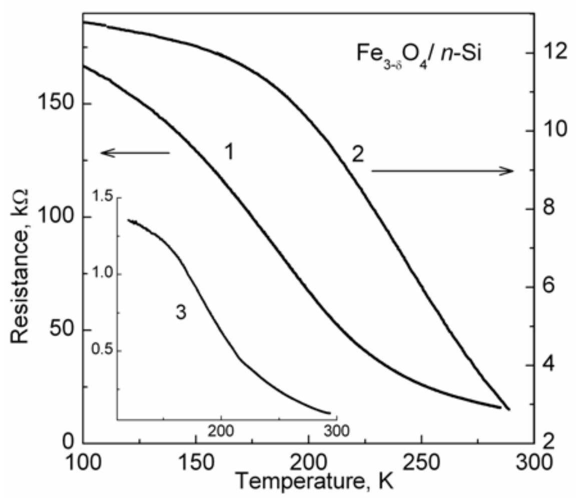

Fig. 1. The interface resistance vs temperature plots measured the $Fe_{3-\delta}O_4$/*n*-Si heterostructures with three different $Fe_{3-\delta}O_4$ layers: *1*- nonstoichiometric $Fe_{3-\delta}O_4$ exhibiting oxygen excess, *2*- stoichiometric Fe_3O_4, and *3* – stoichiometric Fe_3O_4 containing negligible amount of Fe clusters.

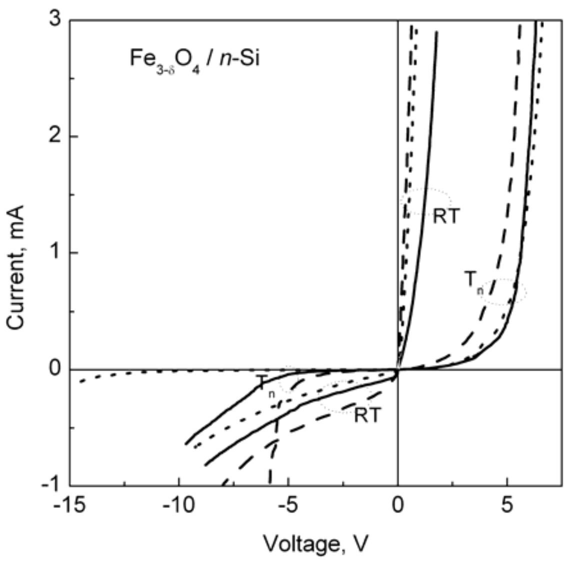

Fig.2. The current-voltage characteristics measured at T = 300 K and 78 K for the $Fe_{3-\delta}O_4$/*n*-Si heterostructures with three different $Fe_{3-\delta}O_4$ layers: nonstoichimetric $Fe_{3-\delta}O_4$ with excess oxygen content (doted line), stoichiometric Fe_3O_4 (solid line) and stoichiometric Fe_3O_4 containing Fe clusters (dashed line).

Crystalline structure of the grown films was studied by measuring their Θ-2Θ X-ray diffraction (XRD) spectra, and studying high-energy electron diffraction (RHEED) images while atomic force microscopy (AFM) was employed to study surface quality of the films. Transport properties of the films were investigated in a wide range of temperatures (T = 78÷300 K) by applying four point-probe method. Meanwhile three point-probe method was applied to investigate interface resistance and I-V dependencies of the prepared p-n device structures.

II. Results and Discussion

The magnetite films were patterned by putting mettalic foil mask with a set of opened squares (1 × 1 mm^2) on the substrates prior to film deposition to estimate contact resistance of the interfaces (between p-$Fe_{3-\delta}O_4$ layers and n-Si or underlaying ITO(IO) layers) as well as to study the corresponding current-voltage (I-V) characteristics. The current versus voltage (I-V) dependencies were measured in a wide temperature range (T = 78÷300 K) by passing dc current I=0÷100 μA and using metallic In pads as electrodes.

$Fe_{3-\delta}O_4$/n-Si heterostrutures. Figure 1 demonstrates the interface resistance versus temperature for three different $Fe_{3-\delta}O_4$/Si heterostructures. Curve 2 in this figure corresponds to the heterostructure with stoichiometric Fe_3O_4 layer while curves 1 and 3 were measured for similar heterostructures containing magnetite layers with excess oxygen content (film with Fe_2O_3 inclusions) and oxygen deficiency (with negligible amount of mettalic Fe clusters), respectively.

Figure 2 demonstrates current-voltage (I-V) dependencies measured for the same heterostructures at T = 300 K and 78 K. Clearly defined rectifying behavior typical for semiconductor p-n diode structures have been indicated for the heterostructures both at room temperature and at T = 78 K. It can be seen from Fig. 2 that differential resistance of the heterostructures at zero bias (R_d =dV/dI) increased significantly with cooling down to T = 78 K. In all cases, current through the interface was found to increase steeply with forward bias (V > 0) at a certain critical value V_d (diffusion voltage) corresponding to the mismatch between band structures of p-$Fe_{3-\delta}O_4$ and n-type Si. It leads from Fig. 2 that the heterostructure with enhanced oxygen content in the magnetite layer has greater influence on electrical properties of the interface compared to that containing oxygen deficient magnetite layer (see doted and dashed curves in the Figure 2, respectively,).

$Fe_{3-\delta}O_4$/(IO)ITO heterostrutures. Relatively low interface resistance and almost linear current versus voltage dependencies were measured for the $Fe_{3-\delta}O_4$/ITO heterostructures in the whole temperature region (T = 78÷300 K). Presence of intermediate IO layer with reduced carrier density (compared to that of ITO layer) resulted significant increase of interface resistance and revealed nonlinear properties of the heterojunctions. The interface resistance versus temperature plots measured for two different Fe_3O_4/IO(ITO) heterostructures are presented in Figure 3. Curve 1 in this figure corresponds to the heterostructure containing stoichiometric Fe_3O_4 overlayer while curve 2 was measured for similar heterostructure containing non-stoichiometric (oxygen deficient) magnetite layer.

Figure 4 demonstrates current-voltage (I-V) dependencies measured at T = 78 K for a set of $Fe_{3-\delta}O_4$/(IO)ITO heterostructures containing $Fe_{3-\delta}O_4$ overlayers with different Fe/O ratios (different oxygen content). In all theese cases, current flowing through the IO/ $Fe_{3-\delta}O_4$ interfaces increased nonlinearly with bias voltage (V) increasing. Significant nonlinearity and

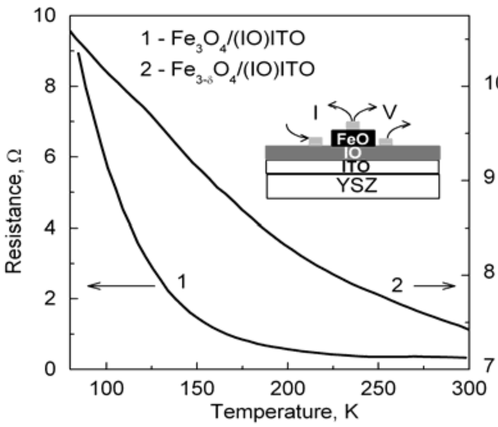

Fig. 3. The interface resistance versus temperature measured for the $Fe_{3-\delta}O_4$/(IO)ITO heterostructures containing stoichiometric Fe_3O_4 overlayer (1) and that containing Fe clusters (2).

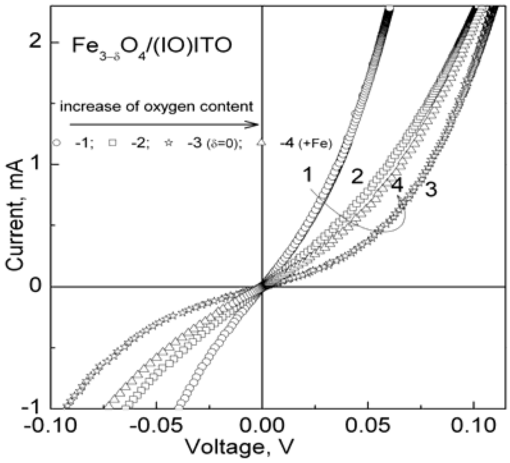

Fig. 4. The *I-V* characteristics measured at *T*=78 K for the $Fe_{3-\delta}O_4$/(IO)ITO heterostructures with three different $Fe_{3-\delta}O_4$ layers: *1, 2* - nonstoichiometric $Fe_{3-\delta}O_4$ exhibiting oxygen excess, *3*- stoichiometric Fe_3O_4, and *4* – stoichiometric Fe_3O_4 containing Fe clusters.

clearly defined asymmetry of the I-V curves (in a case of forward and reverse bias) were indicated for the heterostructure with stoichiometric magnetite layer (see curve 3) while reduced rectifying properties were indicated for the heerostructures with nonstoichiometric $Fe_{3-\delta}O_4$ layers. It is worth noting also that slight non-linearity of the I-V characteristics were also seen at low temperatures above the characteristic Verwey transition temperature ($T_v \sim 120$ K). However, almost linear I-V dependencies were measured for all the prepared $Fe_{3-\delta}O_4$/(IO)ITO heterostructures at room temperature. The observed linear I-V characteristics of the $Fe_{3-\delta}O_4$/(IO)ITO heterostructures at 300 K may be understood assuming tunneling of carriers through the interface between highly conducting IO and $Fe_{3-\delta}O_4$ layers. Significant resistance increase observed for magnetite at $T \cong T_V$ (the Verwey transition temperature) have been associated to charge ordering and occurance of energy gap in the energy spectrum of Fe_3O_4. Nonlinearity and asymmetry of the I-V curves of the $Fe_{3-\delta}O_4$/(IO)ITO heterostructures at $T < T_V$ may be explained taking into account creation of a depletion region due to reduced carrier concentration at the interface similar to a typical semiconductor p-n junction.

Summarizig

We conclude that p-n heterostructures containing p-type Fe_3O_4 layers on n-type Si substrates demonstrate rectifying behaviour in a wide temperature range (T = 78÷300 K) while those prepared by growing Fe_3O_4 layers on indium oxide (IO) demonstrate nonlinear current-voltage (I-V) dependencies only at low temperatures (T < 120K).

This work has been supported partially by the Swedish government Visby program and Lithuanian State Science and Studies Foundation (Grant No T-102/07).

[1] M. Ziese. Extrinsic magnetotransport phenomena in ferromagnetic oxides// *Rep. Prog Phys*. **65**, pp. 143–249 (2002).

[2] M. Ziese,U. Köhler, A. Bollero, R. Höhne, and P. Esquinazi. Schottky barrier and spin polarization at the Fe_3O_4-Nb:$SrTiO_3$ interface// *Phys Rev B* **71**, p. 180406(R) (2005).

[3] Darshan C. Kundaliyaa, S. B. Ogale, L. F. Fu, S. J. Welz, J. S. Higgins, G. Langham, S. Dhar, N. D. Browning, T. Venkatesan. Interfacial characteristics of a Fe_3O_4/Nb(0.5%):$SrTiO_3$ oxide junction// *JAP*, p. 08K304 (2006).

В. Лісаускас, Б. Венгаліс, К. Шлюжене, Р. Буткуте

Нелінійні електричні властивості p-n гетеро структур на основі феромагнетика Fe_3O_4

Інститут фізики напівпровідників, вул.. А. Гоштауто, Вільнюс, Литва, E-mail: lisa@pfi.lt

У роботі підготовлено і досліджено p-n гетероструктури, які складаються з шарів феромагнетика Fe_3O_4 p-типу вирощених на підкладках Si n-типу, шарів оксиду індію (OI) n-типу, а також оксиду індію легованого оловом(IOO). Плівки Fe_3O_4 (d = 60-600 нм) осаджували магнетронним розпиленням за температури T = 300-450 ºC. Тонкі плівки оксиду індію та оксиду індію легованого оловом товщиною (d = 200-500 нм) приготовлені на підкладки YSZ(100) за температури 250-600 ºC розпиленням на чистий індій або на сплав In-Sn (91:9). Гетероструктури Fe_3O_4/n-Si демонструють нелінійну вольт-амперну характеристику (I-V), яка має залежність в діапазоні температур (T = 78-300 K), а для гетероструктур Fe_3O_4/(OI)IOO показана асиметрія та нелінійність тільки за температури T < 120 K.