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Low-temperature conducting channel switching in hybrid $Fe_3O_4/SiO_2/n$-Si structures

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ABSTRACT

The carrier transport properties of the polycrystalline magnetite (Fe_3O_4) films grown on an *n*-type Si substrate with 5 nm-thick SiO_2 have been investigated between 80 and 300 K in current-in-plane geometry. It was established that at temperature decrease to about 120 K, the resistivity of thin Fe_3O_4 films increases up to a peak value and then abruptly drops. This process is attended by a change in the shape of the current-voltage characteristics from the linear at 300 K to the *S*-type at 80 K. The observed peculiarities are explained by conducting channel switching from the Fe_3O_4 film to the Si substrate via the field-assisted tunneling of carriers through the composite insulating layer consisting of highly resistive Fe_3O_4 and tunnel SiO_2.

1. Introduction

Silicon metal-oxide-semiconductor (MOS) structures are very attractive since they constitute the basis of an MOS field-effect transistor [1]. The transport properties of such structures are traditionally studied in geometry with the current directed perpendicular to the planes of interfaces. This geometry is relatively simple for a theoretical analysis and interpretation of experimental results. However, current-in-plane (CIP) geometry, for cases when a current flows along the interfaces, is sometimes preferable for practical purposes. One of the most notable effects associated with carrier transport in CIP geometry is conducting channel switching (CCS) between conductive film and semiconductor substrate [2–5]. This effect shows up in the temperature dependence of the resistivity (ρ(T)) of conductive films formed on semiconductor substrates with a tunnel insulator and manifests itself as a sharp decrease of the measured value when temperature rises in the interval 200–300 K. According to Dai et al. [2], in the metal-SiO_2-Si structure the decrease of resistivity results from the emission of carriers from the metallic film into the Si inversion layer at the SiO_2/Si interface by thermal excitation. Effect of CCS is well studied in silicon MOS structures with a native oxide and films of metal (Cu, Co [2], Fe [3]) or binary compounds (FeSi, CoSi, Fe_2O_3 [4], Fe_3C [5]), which are characterized by the weak temperature dependence of the resistivity. Contrary to the aforesaid materials, the resistivity of half-metallic magnetite increases by a factor of 10^5 with decreasing of temperature from 300 K to 80 K [6]. However, CCS effect in silicon MOS structures with magnetite films is scantily investigated and data are limited to only a few papers [7–10].

It should be noted that CCS can be characterized by two parameters: the switching temperature and the switching amplitude. The switching temperature is defined as the middle point between the high and low resistance states and can be controlled by the value of the bias current [5,7]. Thus, for the Fe_3C/SiO_2/Si structure the switching temperature shifts from 270 to 300 K with current increase from 10 to 1000 μA [5], whereas for the $Fe_3O_4/SiO_2/n$-Si structure the switching temperature shifts from 220 to 260 K at a similar current alteration [7]. The switching amplitude ($\Delta\rho$), which can be defined as the difference between the resistance values before and after the switching, depends on the type of substrate, its doping level, and film thickness. Results of many studies [2–5,7] indicate that the resistance can change by several tens percent or by several orders of magnitude in the case of the highly conductive films of single-component materials and in the case of the poorly conductive films of binary compounds, respectively. A more pronounced change in $\Delta\rho$ can be the result of a variation of the film thickness. As shown in Ref. [11], $\Delta\rho$ decreases in five times of magnitude down to the complete disappearance of CCS with increase Co film thickness from 3 to 44 nm in a Co/SiO_2/Si structure. This clearly demonstrates that the CCS in silicon MOS structures with tunnel SiO_2 can occur only in a limited range of film thicknesses. For the hybrid structures with magnetite film, the data on the thickness dependence of the switching amplitude differ essentially from each

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Abbreviations: CCS, conducting channel switching; CIP, current-in-plane.
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other. Thus, Zervos et al. [10] have studied the thermoelectric properties of silicon MOS structures with 150 and 200 nm magnetite films grown by PLD on *p*-type Si with native SiO_2 and have found that the switching amplitude in such structures constitutes about two orders of magnitude of the resistivity. However, Wang et al. [7,8] have observed a similar magnitude of $\Delta\rho$ for the samples with thinner (35 and 70 nm) Fe_3O_4 films grown by the laser MBE on *n*-type Si with native SiO_2.

It is known [12] that the magnetite is characterized by a high level of the spin polarization of electrons that makes it a promising material for spintronics. Therefore, the further investigation of CCS in silicon MOS hybrid structures with thin magnetite film should enhance the functionality of spintronic devices. In this paper, we report on the results of an investigation of the carrier transport properties of $Fe_3O_4/SiO_2/n$-Si structures with thicknesses of the polycrystalline magnetite film being less than 70 nm. We demonstrate that the effect of CCS in the structures with magnetite films can also take place at temperatures below 125 K.

2. Experimental procedure

Fe_3O_4 films with thickness of 35, 45 and 70 nm were grown on oxidized Si(001) substrates by the reactive deposition of Fe in an O_2 atmosphere. Details of the growth conditions and parameters are described elsewhere [13]. The *n*-type Si(001) phosphorus doped wafer with a resistivity of 7.5 Ω·cm that corresponds to the doping level of $6 \cdot 10^{14}$ cm^{-3} was used as the substrates. The SiO_2 layers with thickness of ~5 nm were grown on the substrates surface by dry thermal oxidation at 1073 K [14]. SiO_2 thickness was estimated by an iterative fashion using the software Spectroscan supplemented to the spectral ellipsometry complex “Ellipse -1891” [15]. According to reflection high-energy electron diffraction, the formed magnetite films were polycrystalline and had (311)-preferred texture that was identified by the kinematic approach [16]. Using the atomic force microscopy it was determined that the Fe_3O_4 films with thickness of 70, 45 and 35 nm have the grain size equal 45, 30 and 20 nm, respectively. The 70 nm-thick magnetite film grown on a thick (1200 nm) SiO_2 layer for comparison (hereafter, “reference sample”) had the same texture and grain size as in the case of 70 nm-thick film grown on the thin oxide. The carrier transport properties of the samples were studied at temperatures from 80 to 300 K by a standard DC four-probe method in CIP geometry using a KEITHLEY 2400 SourceMeter at a fixed bias current of 1 μA. The resistivity was extracted by taking into account the thickness of Fe_3O_4. The current-voltage (*I-V*) characteristics were taken in a current scanning regime. Current electrodes 0.2 mm in diameter were formed collinearly with a step of 1 mm by thermal evaporation of Al on the Fe_3O_4 film through a metal mask. A schematic diagram of the measurement configuration is shown in the inset of Fig. 1(b).

3. Experimental results and discussion

Fig. 1a shows the temperature dependencies of resistivity of our samples in the linear plot. The resistivity of the “reference sample” (*curve 1*) is 0.07 Ω·cm at 300 K, which is about one order of magnitude smaller than the resistivity of polycrystalline Fe_3O_4 films [17] and is one order of magnitude larger than the resistivity of epitaxial films having similar thickness [18]. One can see that the resistivity of the “reference sample” varies slightly with decreasing temperature from RT to about 150 K, after that it increases sharply and reaches a value of 250 Ohm·cm at 80 K. Such behavior of ρ(T) is typical for polycrystalline Fe_3O_4 films deposited on non-oriented substrates and corresponds to the metal-insulator transition (Verwey transition) in magnetite below 150 K [19]. It was found that reduction of the SiO_2 thickness has a significant impact on the shape of the ρ(T) dependence (*curves* 2, 3, 4). The resistivity of the 70 nm-thick film deposited on the thin SiO_2 (*curve* 2) has a lower than that the “reference sample”, value - 0.04 Ω·cm at T = 300 K. With decreasing temperature the ρ(T) dependence coincides with the one for the “reference sample” down to 110 K. Below 110 K the increase of resistivity slows down and tends to saturation (*curve* 2). For the samples with film thicknesses of 45 nm (*curve* 3) and 35 nm (*curve* 4), the ρ(T) dependencies at low temperatures differ significantly from the one for the sample with 70 nm–thick film (*curve* 2). Namely, with decreasing temperature the resistivity increases up to a peak value and then decreases. Besides, as can be seen in Fig. 1a, the maximal accessible resistivity of 35 and 45 nm-thick Fe_3O_4 films is 50 Ω·cm which is achieved at temperatures of 120 and 110 K, respectively.

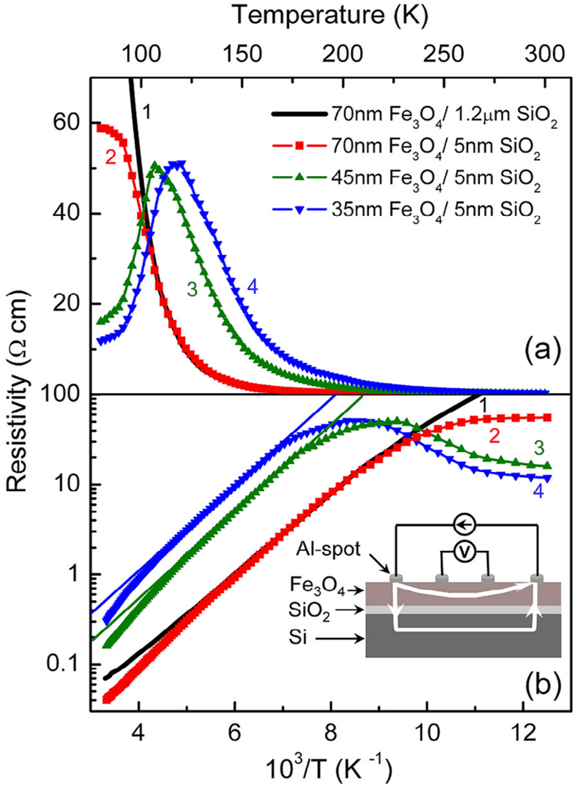

Fig. 1. Temperature dependences of the resistivity of the Fe_3O_4 films with different thicknesses in linear (a) and Arrhenius plot (b). The solid lines in (b) indicate the interpolation of linear part of curves. The inset shows a schematic view of the electrical contact configuration.

Fig. 1b shows the experimental data in Arrhenius coordinates. It is clear that the ρ(1/T) dependence of the “reference sample” (*curve* 1) is linear in the temperature interval of 120–300 K, indicating the thermally activated mechanism of carriers transport. The value of activation energy determined from this curve is 90 ± 3 meV, which is 1.5 times higher than the value found in Ref. [20] for polycrystalline film with thickness of 250 nm, formed on Corning 0211 glass by oxidation of deposited Fe film in an O_2 atmosphere followed by annealing at 673 K. The difference in activation energies may be due to the different methods of films preparation that leads to different structural and morphological properties of the films. For the film thickness of 70 nm (*curve* 2) formed on the thin SiO_2, the ρ(1/T) dependence is also linear in the interval 110–200 K and coincides closely with that of the “reference sample”.

The activation energy was found to be 93 ± 3 meV. Films with thickness of 45 and 35 nm have the same value. The similarity of the activation energy values for the samples with thin SiO_2 and the "reference sample", suggests that the conductivity of the Fe_3O_4/SiO_2/*n*-Si structure in this interval is determined by the properties of the magnetite film alone. As one can see in Fig. 1b, at high and low temperatures the linearity of $\rho(1/T)$ dependence for the 70 nm film (*curve* 2) formed on the thin SiO_2 is no longer fulfilled. For thinner Fe_3O_4 films (*curves* 3, 4), the deviations from linearity become more pronounced. The values of resistivity resulting from extrapolation of the linear interval to 300 K are higher than experimental one. When the temperature decreases, this difference begins to reduce and disappears approximately at 225 K. This temperature is very close to the one of CCS between metal films and silicon substrate in Metal/SiO_2/Si [2,5,21] and Fe_3O_4/SiO_2/Si [7] structures. We suppose that the observed deviation of the resistivity from the exponential function in high temperature interval is also associated with CCS in our structures due to the emission of electrons by thermal excitation. The total disappearance of deviation with temperature decrease is explained by complete attenuation of thermal excitation.

The shape of $\rho(1/T)$ dependencies of the samples with thin SiO_2 at temperature below ~120 K (i.e. below the Verwey transition) is of special interest. As one can see in Fig. 1b, the $\rho(1/T)$ dependence begins to deviate from linearity upon reaching the same resistivity of 25 Ω·cm. For the sample with the 70 nm film, further temperature decreasing leads to the attainment of saturation of $\rho(1/T)$ dependence at $\rho = 60$ Ω·cm. For the samples with 35 nm and 45 nm films, the resistivity reaches 50 Ω·cm, then begins to decrease and reaches a constant value. Absence of such effect for the "reference sample" clearly indicates that this is not an artifact of the measurements, but associated with a change of carrier transport mechanism in these structures because of the reduced SiO_2 thickness.

In order to understand the revealed features of the carrier transport mechanism in the Fe_3O_4/SiO_2/*n*-Si structures with thin SiO_2, we speculate that in such structures there are always two competing conduction channels – Fe_3O_4 film and silicon substrate. In the case of the presence of a tunnel SiO_2 layer, it is possible to realize conditions under which the conductivity will be carried out through the substrate. Since the tunneling is the most-common conduction mechanism through insulators under high fields in MOS structures [22], we propose the following explanation of the observed peculiarity in the low-temperature interval. At the achievement of the peak resistance with temperature decrease the CCS in the Al/Fe_3O_4/SiO_2/*n*-Si structure is carried out by the field-assisted tunneling of carriers across thin SiO_2 after their passing through the transversal resistance of Fe_3O_4. The absence of revealed features in the $\rho(1/T)$ dependencies for the Metal/SiO_2/Si structures [2,5,21] suggests that they may be related to the lack of the strong temperature dependence of metal resistivity in comparison with magnetite. This is to say that the film becomes almost an insulator due to the Verwey transition that radically changes the distribution of fields and charge states in our structures.

Therefore, at a fixed bias current an increase of the film resistance increases a voltage drop between the contacts on the film under current electrodes. This leads to redistribution of carriers in substrate layer adjacent to the SiO_2. As a result, the bias voltage on the thin SiO_2 under the current electrodes increases that causes an increase of probability of carriers tunneling through the potential barrier into the substrate. Thus, in addition to the current through the film, the current across composite insulating layer comprised of transversal highly-ohm Fe_3O_4 resistor and tunnel SiO_2 begins to pass into the substrate (see inset in Fig. 1b). This results in deviation from linearity of $\rho(1/T)$ dependencies in the low temperature range (Fig. 1b). The increasing of film resistivity with the temperature decreasing will lead to increase of the bias voltage up to a critical value. At this value, the electric field in SiO_2 becomes sufficient for the appreciable emission of carriers from current electrodes through Fe_3O_4 via field-assisted tunneling across SiO_2 into the substrate. When the longitudinal resistance of Fe_3O_4 film becomes comparable with the resistance of the composite insulating layer, current path is switched from the highly-resistive magnetite film to the low-resistive substrate layer which is adjacent to the SiO_2 (see inset in Fig. 1b). Further increasing of film resistance with temperature will increase the bias voltage and enhance the field-assisted tunneling of carriers into the substrate. That is the reason why we observe the maxima on the $\rho(1/T)$ dependencies in low temperature interval.

It is remarkable that the critical value of voltage depends on the transversal resistance of Fe_3O_4 film, which, in turn, is determined by the temperature and the film thickness. Therefore, for the structure with 35 nm-thick Fe_3O_4 film the switching temperature is 117 K, whereas, for the structure with 45 nm-thick Fe_3O_4 film it is lower - 107 K. For these samples, when the bias voltage reaches the critical value, CCS occurs, and the measured resistivity decreases sharply. For the structure with a film thickness of 70 nm, the critical value of voltage is not reached since the longitudinal resistance of thick magnetite film is low. Thus, it can be supposed that CCS in the low temperature interval may occur only for a limited range of magnetite film thicknesses.

For a more complete understanding the mechanism of carrier transport in hybrid Al/Fe_3O_4/SiO_2/*n*-Si structures, the current-voltage characteristics were measured at 300 K and 80 K (Fig. 2). We observed the clear linear relationship between current and voltage at room temperature (Fig. 2a) that is typical for half-metallic films of magnetite [19]. With a decrease of film thickness, the slope of *I-V* curves decreases, indicating an increase of resistance. The linearity of *I-V* curves at 300 K and their dependence on the film thickness may signify that prevalent conduction channel in these structures is located in the magnetite films. When the conductivity is caused by the channel in silicon substrate, the *I-V* characteristics of Metal/Fe_3O_4/SiO_2/*n*-Si have a nonlinear shape [8].

As one can see in the inset of Fig. 2b, at 80 K, the *I-V* characteristic of the "reference sample" has an initial region of low conductivity. After reaching +/- 0.5 V, a steep increase of current takes place followed by a monotonic increase. Such behavior of *I-V* characteristic is explained by the opening of the Coulomb gap or an insulating gap in magnetite film [19]. For samples with thin SiO_2, the *I-V* characteristics at 80 K have an *S*-shape with negative resistance region under forward and reverse bias conditions (Fig. 2(b)). Such characteristics are typical for the Metal/Insulator/Semiconductor tunnel structures [23].

To clarify such shape of *I-V* curves, let us consider the energy band diagram of the Fe_3O_4/SiO_2/*n*-Si structure taking into account the work functions difference between Fe_3O_4 and Si, $\chi_{MS} = \chi_{Fe3O4} - \chi_{Si}$ (Fig. 3). In thermal equilibrium with values of $\chi_{Fe_3O_4} = 5.2$ eV [24] and $\chi_{Si} = 4.33$ eV for the doping level *n*-Si of $6 \cdot 10^{14}$ cm^{-3} [22], the value of χ_{MS} is positive and equals to 0.87 eV. This will produce an upwards band bending at the SiO_2/Si interface leading to the formation of an inversion layer even with the absence of applied voltage. The presence of an inversion layer provides a redistribution of the applied voltage, resulting in the positive feedback necessary for the appearance of a region of negative differential resistance in the *I-V* characteristics. We suppose that at low temperature the Al/Fe_3O_4/SiO_2/*n*-Si structure is similar to the MOS tunnel diode, which distinguishes oneself by *S*-shaped *I-V* characteristic [23,25–27]. The Fe_3O_4 film in our structure acts as a current-limiting resistor connected in series with the tunnel diode. The presence of this resistor in an electrical circuit causes the electrically induced bistable state [26] of our structure.

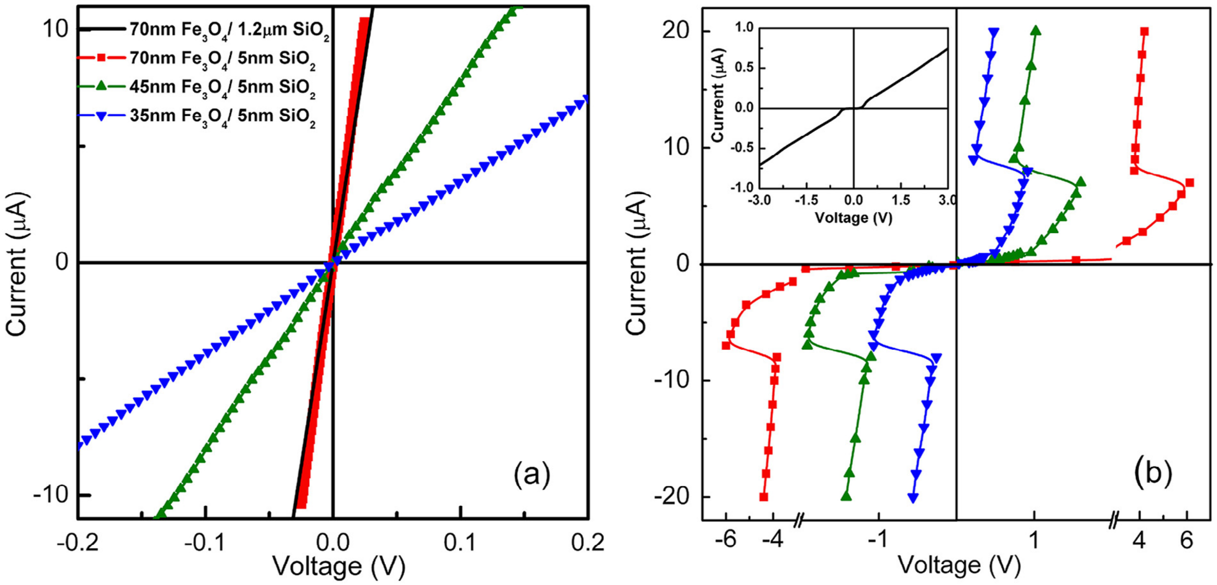

Fig. 2. The *I-V* characteristics of the hybrid Al/Fe_3O_4/SiO_2/*n*-Si structure measured in CIP geometry: (a) at 300 K and (b) at 80 K. The inset in (b) shows the *I-V* characteristics of the “reference sample” at 80 K.

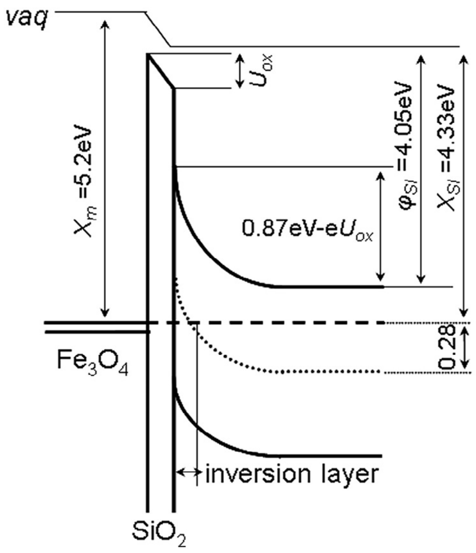

Fig. 3. Energy band diagram of the Fe_3O_4/SiO_2/*n*-Si structure in thermodynamic equilibrium state.

4. Conclusion

In conclusion, we have investigated the carrier transport properties of planar polycrystalline Fe_3O_4/SiO_2/*n*-Si structures with different thicknesses of Fe_3O_4 films in the temperature interval $80 < T < 300$ K. It was established that in addition to the effect of conducting channel switching between the polycrystalline Fe_3O_4 film and the inversion layer of the *n*-Si substrate at a temperature higher than 200 K, a similar effect can take place at temperature below 125 K. It was shown that in contrast to the high-temperature switching provided by thermal excitation of carriers at low temperature it occurs via the field-assisted tunneling through the composite insulating layer that consists of the highly resistive Fe_3O_4 and the tunnel SiO_2. It was demonstrated that the effect of low temperature conducting channel switching is observed in a limited range of Fe_3O_4 film thicknesses (35–70 nm) and disappears at higher thicknesses.

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