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Xiang Liu,¹ Wenbo Mi,^{1,a)} Qiang Zhang,² and Xixiang Zhang^{2,a)}

AFFILIATIONS

¹Tianjin Key Laboratory of Low Dimensional Materials Physics and Preparation Technology, School of Science, Tianjin University, Tianjin 300354, China

²PSE Division, King Abdullah University of Science and Technology (KAUST), Thuwal 23955-6900, Kingdom of Saudi Arabia

^{a)}Author to whom correspondence should be addressed: miwenbo@tju.edu.cn and xixiang.zhang@kaust.edu.sa

ABSTRACT

The electronic transport and magnetotransport properties of $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ heterostructures were investigated with a current source. Negative differential resistance is observed in $\text{Fe}_3\text{O}_4/\text{SiO}_2/p\text{-Si}$ heterostructures. The measurement circuit with four electrodes that $I+$ ($I-$) and $V+$ ($V-$) came into contact with the Fe_3O_4 (Si) layer introduces an in-plane transport into the heterostructures. By decreasing the temperature, the in-plane conductive channel switches from Fe_3O_4 to $p\text{-Si}$. However, the in-plane current is still carried by Fe_3O_4 in $\text{Fe}_3\text{O}_4/\text{SiO}_2/n\text{-Si}$ heterostructures. The formation of an accumulation layer in $p\text{-Si}$ facilitates conductive channel switching (CCS), while the depletion layer in $n\text{-Si}$ hampers the CCS. At 150 K, a magnetic-field-independent magnetoresistance (MR) in $\text{Fe}_3\text{O}_4/\text{SiO}_2/p\text{-Si}$ heterostructures manifests the conductive channel in the space charge region of $p\text{-Si}$. A positive MR generated from the increased electronic scattering in a trapezoidal space charge region reshaped by the magnetic field has been detected.

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Devices with a negative differential resistance (NDR) are appropriate for applications such as oscillators, logic gates, and data storage devices due to their folded current-voltage character.^{1,2} The occurrence of NDR has been reported in many systems including semiconductor quantum wells, superlattices, organic tunneling junctions, metal/oxide heterostructures, and two-dimensional heterostructures.^{3–10} However, the physical mechanisms of NDR are diverse, such as resonance tunneling, redox reaction, and filament formation and rupture.^{6,11–13} Within such complex NDR devices, it is difficult to avoid a complex processing technology during the preparation. Thus, whether the NDR can be realized in a simple way is a prime problem.

The distribution of carriers in the space charge region in metal/oxide/semiconductor (MOS) junctions usually plays an important role in the electronic transport properties of heterostructures. The space charge region can be controlled by the external electric field, and thus, tremendous interest can be stimulated.¹⁴ Previous studies show that the in-plane conductive channel can be switched between the conductive film and the semiconductor substrate in MOS junctions by adjusting the ambient temperature.^{15–18} The potential difference between the conductive film and the semiconductor wafer is thus alternated.

From this point of view, the NDR might exist in this kind of heterostructure at a certain temperature. Fe_3O_4 is an appropriate ferrite which shows an increased resistivity by a factor of 10^5 as temperature decreases from 300 K to 80 K.¹⁹ Specifically, Fe_3O_4 undergoes a first order phase transition at ~ 120 K (T_V), which is known as Verwey transition.^{19–22} Below T_V , Fe_3O_4 is a p -type semiconductor with a bandgap of ~ 0.5 eV.²³ The strong temperature dependence of resistivity may facilitate the occurrence of NDR.

In this letter, the electronic transport and magnetotransport in $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ heterostructures are investigated. The NDR is observed in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/p\text{-Si}$ heterostructure due to the formation of an accumulation layer in $p\text{-Si}$. The magnetotransport investigations further manifest a conductive channel in the space charge region of $p\text{-Si}$.

Polycrystalline Fe_3O_4 films were fabricated by a DC magnetron sputtering apparatus on n - (0.01 Ω cm) and p -type Si (0.02 Ω cm) substrates from a pair of pure Fe (99.99%) targets. The 330- μm -thick Si substrates are covered with a native SiO_2 buffer layer.²⁴ When the base pressure of the chamber was lower than 6.0×10^{-6} Pa, the Ar (100 sccm) and O_2 (4.4 sccm) gas mixture was introduced into the chamber till 1 Pa. The sputtering power on Fe targets was 320 W. The

film thickness of about 170 nm was determined using a Dektak 6M surface profile measuring system. The structure of the $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ sample was determined by X-ray diffraction (XRD) and transmission electronic microscopy (TEM). The temperature dependent Raman spectrum was scanned from room temperature to 87 K. Electronic transport properties of the heterostructures were measured with a current source using a Quantum Design physical property measurement system. Electrodes with four terminals were painted with commercial silver colloidal solution.

Figure 1 shows the structure of the $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ sample. A native 3-nm-thick SiO_2 layer covers the Si substrate. A polycrystalline Fe_3O_4 film with grains can be observed in the low-magnification TEM image [Fig. 1(a)]. The average grain size in the transverse direction is about 290 nm. The XRD results in the inset of Fig. 1(b) indicate that no other phases appear except Fe_3O_4 and Si. In Fig. 1(b), the consistent selected area electronic diffraction patterns and XRD results [inset of Fig. 1(b)] manifest the polycrystalline structure of the Fe_3O_4 film. The lattice constant of 8.326 Å is calculated with the $\text{Fe}_3\text{O}_4(311)$ peak.²⁵ Figure 1(c) shows the temperature dependent Raman spectrum of the $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ heterostructure. The Raman active peaks at $\sim 524\text{ cm}^{-1}$ come from Si. Raman active modes of E_g , $T_{2g}(2)$, and A_{1g} from Fe_3O_4 show blueshift as temperature decreases. The dashed lines indicate the appearance of Raman active peaks from Fe_3O_4 below T_V . These peaks at around 161 cm^{-1} , 339 cm^{-1} , 384 cm^{-1} , 460 cm^{-1} , and 617 cm^{-1} are in accordance with our previous results.²⁶

FIG. 1. (a) The polycrystalline structure of the $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ sample. (b) Selected area electron diffraction patterns together with the XRD results. (c) Temperature dependent Raman spectrum of the $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ junction.

In order to explore the transport properties of the heterostructures, the current-voltage (I - V) characteristics are investigated. The configuration of the transport measurement circuit is shown in the inset of Fig. 2(b). The forward current refers to a positive current applied to the Fe_3O_4 film. Figure 2(a) shows the I - V curves of the $\text{Fe}_3\text{O}_4/\text{SiO}_2/n$ -Si heterostructure. In the current range of $\pm 0.1\text{ mA}$, the I - V curves are almost linear and symmetric above 200 K, while non-linear and asymmetric below 200 K. In the current range of $\pm 10\text{ mA}$, the I - V curves exhibit a backward diode-like rectifying behavior,^{27,28} as shown in the inset of Fig. 2(a). As temperature decreases below 70 K, the I - V curves show a tendency of symmetry. Figure 2(b) depicts the differential resistance of the $\text{Fe}_3\text{O}_4/\text{SiO}_2/n$ -Si junction in the current range of $\pm 0.1\text{ mA}$. The resistance switching is inconspicuous above 200 K. As temperature decreases from T_V to 70 K, the resistance switching shows a sharper change at a forward current than that at a reverse current.

The transport mechanisms across the $\text{Fe}_3\text{O}_4/\text{SiO}_2/n$ -Si interface are investigated by fitting the I - V curves with space charge limited current (SCLC),²⁹ Schottky emission,³⁰ Poole-Frenkel (PF) emission,^{31,32} and Fowler-Nordheim (FN) tunneling³³ regimes. The I - V correlations can be described as

$$I_{\text{SCLC}} = \frac{9\epsilon_i \mu V^2}{8d^3}, \quad (1)$$

$$I_{\text{Schottky}} = A^{**} T^2 \exp \left[\frac{-q(\phi_B - \sqrt{qV/4\pi\epsilon_i})}{k_B T} \right], \quad (2)$$

$$I_{\text{PF}} = V \exp \left[\frac{-q(\phi_B - \sqrt{qV/\pi\epsilon_i})}{k_B T} \right], \quad (3)$$

$$I_{\text{FN}} = V^2 \exp \left[-\frac{4\sqrt{2m^*}(q\phi_B)^{3/2}}{3qhV} \right], \quad (4)$$

where ϵ_i , μ , V , d , A^{**} , T , k_B , ϕ_B , and m^* represent the insulator permittivity, mobility of carriers, voltage on the insulator, thickness of the insulator, effective Richardson constant, temperature, Boltzmann constant, barrier height, and effective mass, respectively. Figure 2(c) shows the transport mechanism below 200 K at different temperatures and currents. Due to a larger work function of 5.5 eV in Fe_3O_4 , electrons in n -Si flow into Fe_3O_4 spontaneously, resulting in a Schottky barrier at the interface [Fig. 2(d)]. With the decrease in temperature, the carriers are emitted across the interface with the assistance of the electric field at a low current. At a high current, electrons from n -Si transport to the conduction band in Fe_3O_4 at a similar energy level by tunneling [Fig. 2(d)]. At a reverse current, the built-in electric field enhances the band bending of Si at the interface. The applied reverse electric field shifts the conduction band below the Fermi level, which leads to a backward diode behavior [Fig. 2(d)]. The carriers thus transport across the barrier by tunneling, which is in line with the fitting results in Fig. 2(c). The precise fitting parameters like the barrier height and the effective mass cannot be obtained by fittings in the FN tunneling regime because the voltage on the insulator is divided by the in-plane transport in Fe_3O_4 , especially at low temperatures. In the Schottky emission regime, ϕ_B is obtained by fittings, which increases with the increase in temperature (Fig. S1 in the [supplementary material](#)).

FIG. 2. (a) I - V characters in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{n-Si}$ heterostructure. (b) Differential resistance of the heterostructure. (c) The critical current of different transport mechanisms in the heterostructure. (d) Diagrams of band alignment of Fe_3O_4 and n-Si . Red and green lines indicate the forward and reverse currents. The inset of (a) shows the I - V curves of the heterostructure with a current range of ± 10 mA. The inset in (b) shows the circuit configuration.

Figure 3(a) shows the I - V curves of the $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{p-Si}$ heterostructure. The transport mechanism follows ohmic behavior above 200 K (240 K) at a forward (reverse) current [Fig. 3(c)]. As temperature decreases from 240 to 100 K, the heterostructure shows the rectification [Figs. 3(a) and 3(b)]. Figure 3(c) shows the transport mechanism at different temperatures and currents. At a reverse current, the band bending at the interface results in a backward diode behavior like $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{n-Si}$ heterostructure [Fig. 3(d)].^{27,28} As temperature decreases below 100 K, the I - V curves exhibit NDR, i.e., the

differential resistance $\Delta V/\Delta I < 0$. By using the current source, the NDR shows an “S” shape, which is different from the “N” shape NDR with a voltage source. At a forward current, the voltage peak can be observed at 70 K. The $V_{\text{peak}}/V_{0.1 \text{ mA}}$ ratio is about 1.5. At a reverse current, the voltage shows a peak from 90 to 70 K. With the decrease in temperature, the ratio of $V_{\text{peak}}/V_{-0.1 \text{ mA}}$ changes from 1.2 at 90 K to -0.14 at 70 K. Peak-valley ratios of about 1.5 and 1.4 are reported in a lateral insulated gate bipolar transistor and a TiO_2 layer, respectively.^{34,35} Compared to the peak-valley ratio of 10^7 in a voltage-controlled NDR

FIG. 3. (a) I - V characters in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{p-Si}$ heterostructure. (b) Differential resistance of the heterostructure. (c) The critical current of transport mechanisms in the heterostructure. (d) Diagrams of band alignment of Fe_3O_4 and p-Si . Red and green lines indicate the forward and reverse currents.

device,³⁶ the ratios in current-controlled devices are rather low. At 60 K, the differential resistance is totally negative. In previous results, the NDR has never been found in $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ heterostructures in a longitudinal two-probe manner.^{27,28,37} So the NDR may derive from the in-plane transport in the heterostructure. As temperature decreases, the Verwey transition increases the resistivity of Fe_3O_4 . In addition, due to the smaller work function, the electrons in p -Si flow into Fe_3O_4 spontaneously, resulting in a depletion layer in Fe_3O_4 [Fig. 3(d)]. Hence, the lateral conductance channel switches from Fe_3O_4 to p -Si at low temperatures due to the increased resistivity of Fe_3O_4 . The onset of CCS decreases the potential difference between $V+$ and $V-$ electrodes. As a result, the I - V curves show NDR. In the $\text{Fe}_3\text{O}_4/\text{SiO}_2/n$ -Si heterostructure, the formation of a depletion layer increases the resistivity of the space charge region, which hampers CCS. The NDR is observed in a $\text{Fe}_3\text{O}_4/\text{SiO}_2/p$ -Si heterostructure with common in-plane geometry, which results from the CCS between Fe_3O_4 and p -Si (Fig. S2 in the supplementary material). In Fig. 3(a), the I - V curves show irreversible inflection below 150 K at a 0.1 mA current. The special behavior is also observed in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/n$ -Si heterostructure at 30 K [Fig. 2(a)] and $\text{Fe}_3\text{O}_4/\text{SiO}_2/p$ -Si heterostructure with in-plane geometry below 120 K [Fig. S2(b)], which may be from the CCS. At a higher temperature, holes accumulate in the space charge region of Si. At the first stage of the measurement, the rapid increased resistivity of Fe_3O_4 at lower temperature facilitates the occurrence of CCS, resulting in a smaller potential difference.

In order to demonstrate the speculation of CCS, the spin-dependent transport in the heterostructures is investigated. Here, the magnetoresistance (MR) of the heterostructure is defined as $\text{MR} = (R_H - R_0)/R_0 \times 100\%$, where R_H and R_0 are the resistance of the junction with and without the magnetic field. In Fig. 4(a), the temperature dependence and the magnitude of MR in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/n$ -Si heterostructure above 240 K are in accord with MR in a polycrystalline Fe_3O_4 film on a quartz substrate,^{24,38} which manifests that the in-plane current is mainly carried by the Fe_3O_4 layer. However, the intrinsic temperature dependence of MR in polycrystalline Fe_3O_4 cannot be reproduced in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/p$ -Si heterostructure [Fig. 4(b)].

Thus, a positive MR may exist in $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ heterostructures. Previous studies show that the rectangular space charge region transforms to a trapezoidal one under a magnetic field, as shown in Fig. 4(d). The electric field inhomogeneity in the space charge region generates a positive MR.^{39,40} With the decrease in temperature, the increased carriers' mobility aggravates the spatial distribution of the space charge region, which results in a decrease in MR. Figure 4(c) shows the magnetic field dependence of MR in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/p$ -Si heterostructure at 150 K. The MR shows a strong dependence on current, which is almost independent of the magnetic field. In Fig. 3(c), at 150 K, the I - V correlation in the current range from -0.0156 to 0.0234 mA is ohmic, suggesting the inconspicuous interface effects on the transport. The temperature of 150 K is a critical temperature of NDR [Fig. 3(a)], which signifies the occurrence of conductive channel switching. Thus, the in-plane current is carried by a Fe_3O_4 layer at a low current while by p -Si at a high current due to the bending band under a high external electric field. The field-dependent MR at low current results from the intrinsic properties of Fe_3O_4 . In the lower panel of Fig. 4(d), at a high current, the redistribution of carriers in the space charge region increases the resistivity of Fe_3O_4 , which leads to an increased critical temperature of NDR. The carriers in Fe_3O_4 are spin-polarized,^{41,42} and so MR may come from the spin accumulation in the space charge region of p -Si.^{43,44} Due to the nonmagnetic essence of p -Si, the accumulated spin-polarized carriers align to the applied magnetic field. The resistivity in the space charge region of p -Si thus shows reduction and no longer increases with the increase in the magnetic field. As a result, the MR at a high current is a competition of the two parts containing a negative MR generated by the spin-dependent transport in Fe_3O_4 and Si and a positive MR produced by the increased scattering in the trapezoidal space charge region. In Figs. 4(a) and 4(b), the positive MR gradually dominates the MR of the junction at a low temperature, where the I - V curves of the heterostructures show nonlinearity until the NDR occurs.

In summary, the NDR is observed in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/p$ -Si heterostructure. The NDR results from the in-plane conductive channel switching between Fe_3O_4 and the accumulation layer of p -Si.

FIG. 4. Temperature dependence of MR in (a) $\text{Fe}_3\text{O}_4/\text{SiO}_2/n$ -Si and (b) $\text{Fe}_3\text{O}_4/\text{SiO}_2/p$ -Si heterostructures under a 50-kOe field. (c) Magnetic field dependence of MR in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/p$ -Si junction at 150 K. (d) Diagram of the migration of carriers under the magnetic field in $\text{Fe}_3\text{O}_4/\text{SiO}_2/n$ -Si (upper panel) and $\text{Fe}_3\text{O}_4/\text{SiO}_2/p$ -Si (lower panel) heterostructures.

However, the NDR is absent in the $\text{Fe}_3\text{O}_4/\text{SiO}_2/n\text{-Si}$ heterostructure due to the depletion layer formed in $n\text{-Si}$. The critical temperature of NDR may be increased by introducing structural defects into Fe_3O_4 films, fabricating on a conductive substrate or selecting other high spin polarized materials with the enhanced resistivity such as $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$.

See the [supplementary material](#) for the temperature dependence of the barrier height in $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ heterostructures and the current-voltage characters in $\text{Fe}_3\text{O}_4/\text{SiO}_2/\text{Si}$ heterostructures at different temperatures with in-plane geometry.

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