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ABSTRACT

The electronic transport and magnetotransport properties of Fe$_3$O$_4$/SiO$_2$/Si heterostructures were investigated with a current source. Negative differential resistance is observed in Fe$_3$O$_4$/SiO$_2$/p-Si heterostructures. The measurement circuit with four electrodes that I$^+$ (I$^-$) and V$^+$ (V$^-$) came into contact with the Fe$_3$O$_4$ (Si) layer introduces an in-plane transport into the heterostructures. By decreasing the temperature, the in-plane conductive channel switches from Fe$_3$O$_4$ to p-Si. However, the in-plane current is still carried by Fe$_3$O$_4$ in Fe$_3$O$_4$/SiO$_2$/n-Si heterostructures. The formation of an accumulation layer in p-Si facilitates conductive channel switching (CCS), while the depletion layer in n-Si hampers the CCS. At 150 K, a magnetic-field-independent magnetoresistance (MR) in Fe$_3$O$_4$/SiO$_2$/p-Si heterostructures manifests the conductive channel in the space charge region of p-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. 11 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$_3$O$_4$ is an appropriate ferrite which shows an increased resistivity by a factor of 10$^5$ as temperature decreases from 300 K to 80 K. 19,20 Specifically, Fe$_3$O$_4$ undergoes a first order phase transition at $\sim$120 K ($T_V$), which is known as Verwey transition. 19–22 Below $T_V$, Fe$_3$O$_4$ is a $p$-type semiconductor with a bandgap of $\sim$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 Fe$_3$O$_4$/SiO$_2$/Si heterostructures are investigated. The NDR is observed in the Fe$_3$O$_4$/SiO$_2$/p-Si heterostructure due to the formation of an accumulation layer in p-Si. The magnetotransport investigations further manifest a conductive channel in the space charge region of p-Si.

Polycrystalline Fe$_3$O$_4$ 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 $\times$ 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 Fe$_3$O$_4$/SiO$_2$/Si sample was determined by X-ray diffraction (XRD) and transmission electron 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 Fe$_3$O$_4$/SiO$_2$/Si sample. A native 3-nm-thick SiO$_2$ layer covers the Si substrate. A polycrystalline Fe$_3$O$_4$/SiO$_2$/Si heterostructure. The Raman active peaks at $1,339$, $1,384$, and $617$ cm$^{-1}$ come from Si. Raman active modes of $E_g$, $A_{1g}(2)$, and $A_{1g}$ from Fe$_3$O$_4$ show blue shift as temperature decreases. The dashed lines indicate the appearance of Raman active peaks from Fe$_3$O$_4$ below $70$ K, the $I$–$V$ curves show a tendency of symmetry. Figure 2(b) depicts the differential resistance of the Fe$_3$O$_4$/SiO$_2$/Si junction in the current range of $0.1$ mA. The resistance switching is inconspicuous above $200$ K. As temperature decreases from $70$ K 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 Fe$_3$O$_4$/SiO$_2$/Si interface are investigated by fitting the $I$–$V$ curves with space charge limited current (SCLC), Schottky emission, Poole-Frenkel (PF) emission, and Fowler-Nordheim (FN) tunneling regimes. The $I$–$V$ correlations can be described as

$$I_{SCLC} = \frac{q\mu V^2}{8d^3},$$

(1)

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

(2)

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

(3)

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

(4)

where $\epsilon_i$, $\mu$, $V$, $d$, $A^{**}$, $T$, $k_B$, $\phi_B$, and $m^*$ represent the insulator permittivity, mobility of carriers, voltage on the insulator, thickness of the insulator, effective Richardson 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$_3$O$_4$ electrons in $n$-Si flow into Fe$_3$O$_4$ 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 conductance band in Fe$_3$O$_4$ at a similar energy level by tunneling [Fig. 2(d)]. At a reverse current, the build-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$_3$O$_4$, especially at low temperatures. In the Schottky emission regime, $\phi_B$ is obtained by fittings, which increases with the increase in temperature (Fig. S1 in the supplementary material).
Figure 3(a) shows the I–V curves of the Fe$_3$O$_4$/SiO$_2$/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 Fe$_3$O$_4$/SiO$_2$/n-Si heterostructure [Fig. 3(d)]. 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.

Compared to the peak-valley ratio of $10^7$ in a voltage-controlled NDR
The onset of CCS measurement, the rapid increased resistivity of Fe$_3$O$_4$ at lower temperature accumulates in the space charge region of Si. At the first stage of the measurement, the I–V curves show NDR in the Fe$_3$O$_4$/SiO$_2$/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 Fe$_3$O$_4$/SiO$_2$/p-Si heterostructure with common in-plane geometry, which results from the CCS between Fe$_3$O$_4$ 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 Fe$_3$O$_4$/SiO$_2$/n-Si heterostructure at 30 K [Fig. 2(a)] and Fe$_3$O$_4$/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$_3$O$_4$ 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 MR = (R$_{H}$–R$_{0}$)/R$_{0}$ × 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 Fe$_3$O$_4$/SiO$_2$/n-Si heterostructure above 240 K are in accord with MR in a polycrystalline Fe$_3$O$_4$ film on a quartz substrate, which manifests that the in-plane current is mainly carried by the Fe$_3$O$_4$ layer. However, the intrinsic temperature dependence of MR in polycrystalline Fe$_3$O$_4$ cannot be reproduced in the Fe$_3$O$_4$/SiO$_2$/p-Si heterostructure [Fig. 4(b)]. Thus, a positive MR may exist in Fe$_3$O$_4$/SiO$_2$/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. 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 Fe$_3$O$_4$/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$_3$O$_4$ 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$_3$O$_4$. 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$_3$O$_4$, which leads to an increased critical temperature of NDR. The carriers in Fe$_3$O$_4$ are spin-polarized, and so MR may come from the spin accumulation in the space charge region of p-Si. 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$_3$O$_4$ 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 Fe$_3$O$_4$/SiO$_2$/p-Si heterostructure. The NDR results from the in-plane conductive channel switching between Fe$_3$O$_4$ and the accumulation layer of p-Si.
However, the NDR is absent in the Fe$_3$O$_4$/SiO$_2$/n-Si heterostructure due to the depletion layer formed in n-Si. The critical temperature of NDR may be increased by introducing structural defects into Fe$_3$O$_4$ films, fabricating on a conductive substrate or selecting other high spin polarized materials with the enhanced resistivity such as La$_{2/3}$Sr$_{1/3}$MnO$_3$.

See the supplementary material for the temperature dependence of the barrier height in Fe$_3$O$_4$/SiO$_2$/Si heterostructures and the current-voltage characters in Fe$_3$O$_4$/SiO$_2$/Si heterostructures at different temperatures with in-plane geometry.

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