

OPTICAL AND MAGNETIC TUNING OF ELECTRICAL TRANSPORT ACROSS THE INTERFACES OF $\text{Zn}_{0.3}\text{Ni}_{0.7}\text{Fe}_2\text{O}_4/\text{CuPc}$ HETEROSTRUCTURE

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We have fabricated a $p\text{-Si}/\text{Zn}_{0.3}\text{Ni}_{0.7}\text{Fe}_2\text{O}_4(\text{ZNFO})/\text{CuPc}/\text{Au}$ heterostructure and investigated its magnetic field-induced tuning of photocurrent. Under illumination of 660nm laser, photocurrent increases with increase of optical power. With simultaneous application of light and magnetic field, a drastic change in the ratio of photocurrent with magnetic field ($I(H)$) to photocurrent without magnetic field ($I(0)$) is observed for optical power variations at a fixed positive bias. A significant decrease in electrical hysteresis width ($|\Delta I|$) with magnetic field at higher optical illumination is noticed.

Introduction

Opto-Spintronic devices possess significant potential for future applications in information technology. This is primarily due to their effective capacity for data storage, high operational speed, and low power consumption^{1,2}. The tunability of photocurrent through magnetic field is an intriguing aspect of these devices. However, one of the main challenges in inorganic-based heterostructures is the less spin diffusion length. To address this issue, it may be possible to utilize an energy-efficient and eco-friendly organic spacer layer in heterostructure³. With this objective, multifunctional inorganic magnetic/organic non-magnetic heterostructures, exhibiting magneto-tunability of photo-current, have been documented in scientific literatures^{4,5}. In this study, we have successfully fabricated a $\text{Zn}_{0.3}\text{Ni}_{0.7}\text{Fe}_2\text{O}_4$ (ZNFO)/CuPc/Au heterostructure on a p-type Si (100) substrate

and investigated its magnetic field-induced tuning of photocurrent. The ZNFO material serves as a spin injector, the organic semiconductor CuPc is expected to act as a carrier transport layer, and the Au capping layer functions as an electrode.

Experimental Details

The synthesis of $\text{Zn}_{0.3}\text{Ni}_{0.7}\text{Fe}_2\text{O}_4$ (ZNFO) nanoparticles was carried out via chemical pyrophoric process⁶. Later, a thin film of ZNFO has been deposited on a p-type Si (100) substrate using the Pulsed Laser Deposition (PLD) technique⁷. Subsequently, a CuPc thin film was deposited onto half of the ZNFO film using a thermal evaporation unit under a pressure of 5×10^{-6} mbar, with the deposition rate ranging from 0.1 to 1.2 Å/s. Finally, an Au electrode was deposited onto the top of the CuPc film using an e-beam unit under the same pressure of 5×10^{-6} mbar, with a deposition rate of 0.1 Å/s. The thicknesses of the CuPc and Au films were 1150 Å and 100 Å, respectively. Silver paste and copper wire were used to make electrical contacts on ZNFO and Au layers. The magnetic field and light dependent DC characteristics of the heterostructure were measured using an electrometer (Keysight, B2902A) and a 660 nm red laser light (THOR LABS).

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Results and Discussions

Fig.1 displays the I-V characteristics of the ZNFO/CuPc heterostructure. The arrows in the figure illustrate the direction of voltage changes ($-4\text{ V} \rightarrow 0\text{ V} \rightarrow 4\text{ V} \rightarrow 0\text{ V} \rightarrow -4\text{ V}$) throughout the entire cycle. The top-left inset shows the schematic device structure with connecting terminals. The I-V characteristics of the device exhibit an irreversible nature i.e. electrical hysteresis when the applied voltage is swept along a cycle. The charge trapping at the grain boundary in the ZNFO layer is thought to be the cause of this effect⁸.

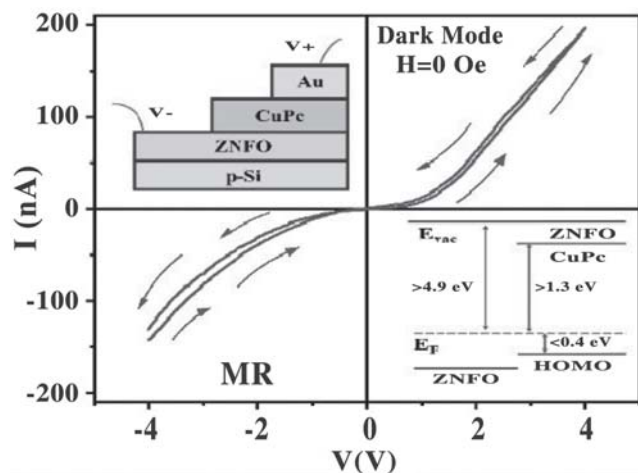


Fig.1. I-V Characteristics of ZNFO/CuPc/Au heterostructure device.

It has previously been documented that hole transport at the ZNFO-CuPc interface is more prominent than electron transport^{9,10}. This phenomenon can be attributed to the relatively small difference in band energy between the highest occupied molecular orbital (HOMO) of CuPc ($<0.4\text{ eV}$) and the Fermi energy level (E_F) of ZNFO, along with the larger band energy difference between the lowest unoccupied molecular orbital (LUMO) of CuPc ($>1.3\text{ eV}$) and the E_F of ZNFO at equilibrium^{9,10}. This observation is illustrated in the energy band diagram of bottom-right inset of Fig.1.

We have studied magneto resistance $\left(MR = \frac{R(H) - R(0)}{R(0)} \right)$ as a function of Magnetic field (H), where $R(H)$ and $R(0)$ represent the device resistance at H and 0 magnetic fields respectively. The MR versus H plot for the device is illustrated in Figure 2. The bottom-right inset of Fig.2 shows the direction of the field with the device. In this plot, a positive MR is evident. It is noticed that MR increases with the increase of magnetic field. At a magnetic field of 3 kOe, a MR of up to 7.15% is observed at a bias of 3.75 V. The use of silver contact (Ag) at the

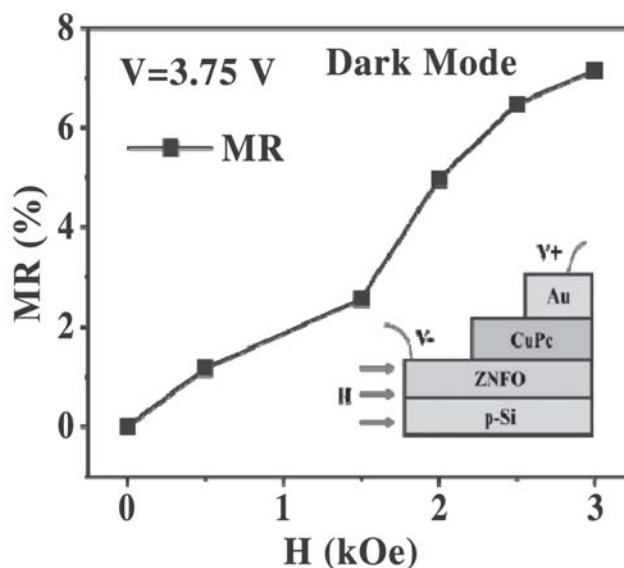


Fig.2. MR versus Magnetic field plot under dark condition.

electrode/ZNFO interface suggests Ohmic contact, resulting in minimal spin-dependent scattering. Similarly, carriers are expected to experience minimal spin-dependent scattering while traversing the ZNFO active region. Organic/inorganic interfaces typically exhibit a high degree of spin polarization at room temperature¹¹. Therefore, the presence of positive MR at low bias at room temperature indicates a high degree of positive spin polarization at the ZNFO/CuPc interface¹⁰.

Fig.3 shows the I-V characteristics of ZNFO/CuPc heterojunction under dark and red light (660 nm) illumination of different intensities. When the device is subjected to light illumination with a wavelength of 660 nm laser, a significant photo response is observed. At a

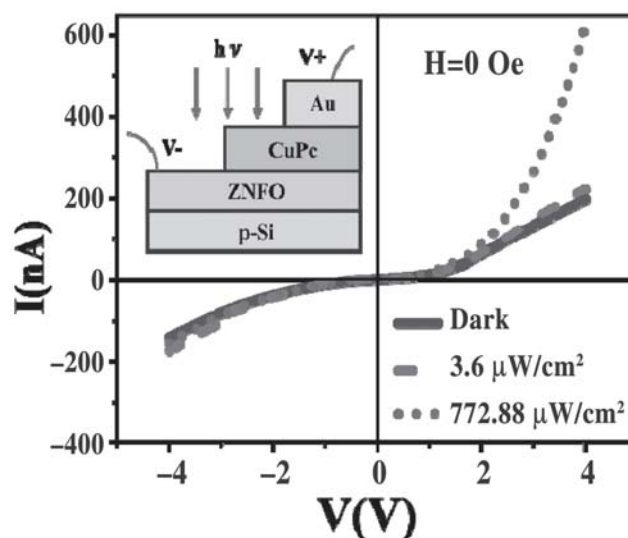


Fig.3. I-V characteristics of the device under dark and red light (660 nm) illumination of different intensities.

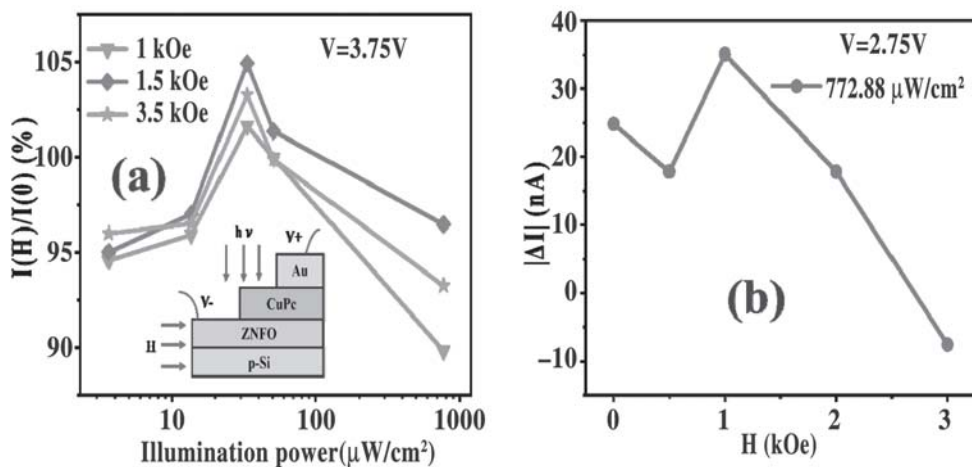


Fig.4. (a) $I(H)/I(0)$ versus Illumination power plot for different magnetic field at 3.75 V. (b) electrical hysteresis width (ΔI) versus Magnetic field (H) plot for illumination power of $772.88 \mu\text{W}/\text{cm}^2$ at 2.75 V.

very low power intensity of $3.62 \mu\text{W}/\text{cm}^2$, the device current begins to increase in the direction of positive bias. As the optical power is increased to $772.88 \mu\text{W}/\text{cm}^2$, a substantial increase in current is observed, indicating the efficient generation of photocurrent by the device. The efficient photocurrent generation under illumination is attributed to Frankel exciton generation at the CuPc bulk and separation at the ZNFO/CuPc interface in the device¹⁰. To investigate the couple effect of light and magnetic field in this heterostructure, we have plotted $I(H)/I(0)$ versus Illumination power for different magnetic fields at a fixed bias of 3.75 V, as shown in Figure 4 (a). Here $I(H)/I(0)$ = Photocurrent with/without magnetic field. We have observed an increase in the ratio of photocurrent with magnetic field ($I(H)$) to photocurrent without magnetic field ($I(0)$) at lower optical power, followed by a decrease at higher optical power for a fixed positive bias. Figure 4 (b) shows electrical hysteresis width (ΔI) versus Magnetic field (H) plot for illumination power of $772.88 \mu\text{W}/\text{cm}^2$ at a constant bias of 2.75 V. Here $|\Delta I|$ is the absolute current difference between two half cycles at constant bias. We have noticed a significant decrease in $|\Delta I|$ with magnetic field at higher optical illumination. The change in $|\Delta I|$ with the magnetic field could be due to magnetostriction and reorientation of magnetic clusters in the presence of an external magnetic field^{8,12}. These observations indicate a magnetic tuning effect on the photogeneration in this device.

Conclusion

In conclusion, we have fabricated ZNFO/CuPc/Au hetero structure device on a p-type Si (100) substrate and studied its light and magnetic field dependent DC transport

properties. The I-V response suggests carrier tunnelling through ZNFO/CuPc, which is mainly due to hole transport across the heterostructure. Under application of magnetic field, a positive MR is observed at forward bias voltages, which can be due to high degree of positive spin polarization within the ZNFO/CuPc interface. Under illumination of 660 nm light, the device shows substantial photo response.

However, when both magnetic field and light are applied simultaneously, a drastic change in the ratio of photocurrent with magnetic field ($I(H)$) to photocurrent without magnetic field ($I(0)$) is observed for optical power variations at a fixed positive bias. The observation of the photo response that is dependent on the magnetic field, as well as the electrical hysteresis effect that is dependent on both light and magnetic field, holds potential for future investigations in the advancement of Opto-Spintronic sensor devices.

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