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ARTICLE

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TUNING OF I-V CHARACTERISTICS OF HYBRID INORGANIC-ORGANIC p-Si/ $La_{0.7}$ $Sr_{0.3}MnO_3$/CuPc/Au HETEROSTRUCTURE UNDER EXTERNAL PERTURBATIONS OF LIGHT AND MAGNETIC FIELD

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We investigated the p-Si/$La_{0.7}$ $Sr_{0.3}$ MnO_3/CuPc/Au hybrid heterostructure under red light and magnetic field. Under dark condition, it displayed a nonlinear, irreversible I-V curve, likely due to the Schottky nature of the LSMO/CuPc heterojunction. The device exhibited positive MR, which may arise from increased spin-dependent scattering at the LSMO depletion region. Increase in light intensity likely boosted exciton generation and dissociation at the LSMO/CuPc interface, enhancing photocurrent through the device.

Introduction

Lately, Opto-Spintronics has emerged as an intriguing branch of Spintronics, merging the high operational speed of Opto-electronics with the advantageous characteristics of Spintronics, such as low power consumption and efficient data manipulation. This convergence has led to multifunctional spintronic devices capable of swiftly and accurately controlling electron spins. Prior studies in Opto-Spintronics have investigated thin films and heterostructure devices, yielding significant findings. L. Ren et al. demonstrated an increase in photocurrent in perovskite solar cells through the application of a magnetic field[1]. B. Luo et al. observed a significant magneto-tunable photocurrent in LSMO/BSO p-n heterojunctions[2], while S. Singh et al. achieved magnetic tuning of photocurrent in $CoFe_2O_4$ nanostructures[3]. V.V. Pavlov et.al. showed magnetic field-enhanced photocurrent in SiO_2 hetero structure films containing cobalt nanoparticles on a GaAs substrate at room temperature[4].

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One of the major challenges encountered by spintronic devices is spin relaxation. This led to the emergence of a young field, known as Organic Spintronics. It addresses these challenges by offering properties like low production cost, flexibility, and energy efficiency. The small spin-orbit coupling and hyperfine interaction in these Organic semiconductors (OSCs) weaken spin relaxation, maintaining spin polarization at larger scales[5]. Cinchetti et al. reported strong spin injection efficiency (around 85-90%) at the interface of Co/CuPc heterostructures, indicating a large mean free path in CuPc[6].

Scientists have successfully developed efficient organic spin valves by utilizing spin injection through spin-polarized hybrid states. Spin valves serve as fundamental building blocks for spintronic devices. Our aim is to explore the Opto-spintronics effects on spin valves. To achieve this goal, we investigated the properties of the half spin valve under the influence of both light and magnetic field. A spin valve typically comprises two interfaces: one between the spin injector and organic spacer layer, and the other between the spacer layer and the spin detector. Understanding the coupled effect of magnetic fields and light on such devices is complex. Thus, we studied a half spin valve device p-Si/LSMO/CuPc/Au. Notably, LSMO exhibits properties of a half-metallic ferromagnet, making it an exceptional spin injector/detector with nearly 100% spin polarization at low temperatures. Ahmadi et al. observed an increase in photocurrent in inorganic/organic perovskite solar cells, attributed to a reduction in recombination induced by photogenerated dipoles[7]. Debajit Deb et al. investigated carrier transport properties in ITO/LSMO/CuPc/Au (Device A) and p-Si/ZNFO/CuPc/Au (Device B) heterostructure devices under light and magnetic field influences. Efficient photocurrent generation, attributed to Frankel exciton generation at the CuPc bulk, was observed in Device A. Impedance measurements revealed magnetic field-dependent scattering at the LSMO/CuPc interface and LSMO bulk. A positive magnetoresistance (MR) at low bias in Device B indicated substantial positive spin polarization within the ZNFO/CuPc interface at room temperature[8]. In this context, we fabricated p-Si/LSMO/CuPc/Au hybrid heterostructure half-spin valve device on a Si substrate and explored its carrier transport properties under varying light and magnetic field conditions.

Experimental Details

First part: We fabricated half-spin valve device with one ferromagnetic electrode, structured as p-Si/LSMO/Copper phthalocyanine (CuPc)/Au. LSMO nanoparticles were prepared using a low-temperature chemical pyrophoric reaction process[9]. Thin LSMO films were deposited on p-Si coated glass substrates using PLD[10] technique with a KrF excimer laser at an optimized substrate temperature of 480°C. A laser fluence of 240mJ and a repetition rate of 10Hz were employed for a total deposition time of 1 hour. Post-deposition annealing was performed for 30 minutes under a constant oxygen partial pressure to achieve good surface morphology. CuPc was deposited over the LSMO film using thermal evaporation, followed by Au deposition using electron-beam evaporation under high vacuum conditions.

Second Part: The device was subjected to I-V measurements using a source meter from Key sight (B2902A), both in the absence of light (dark conditions) and under illumination (660 nm red laser). Light-dependent

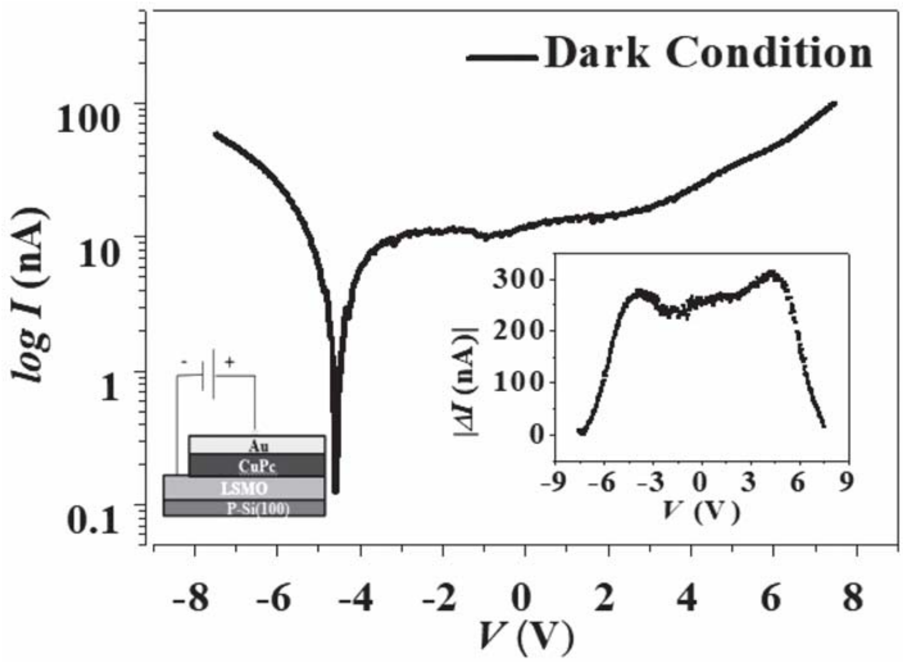

Fig.1. I-V Characteristics of p-Si/LSMO/CuPc/Au heterostructure in the absence of light and magnetic field (Dark Condition). Left bottom inset shows the schematic of biasing of the heterostructure device diagram. The right bottom inset shows the |ΔI| vs V plot.

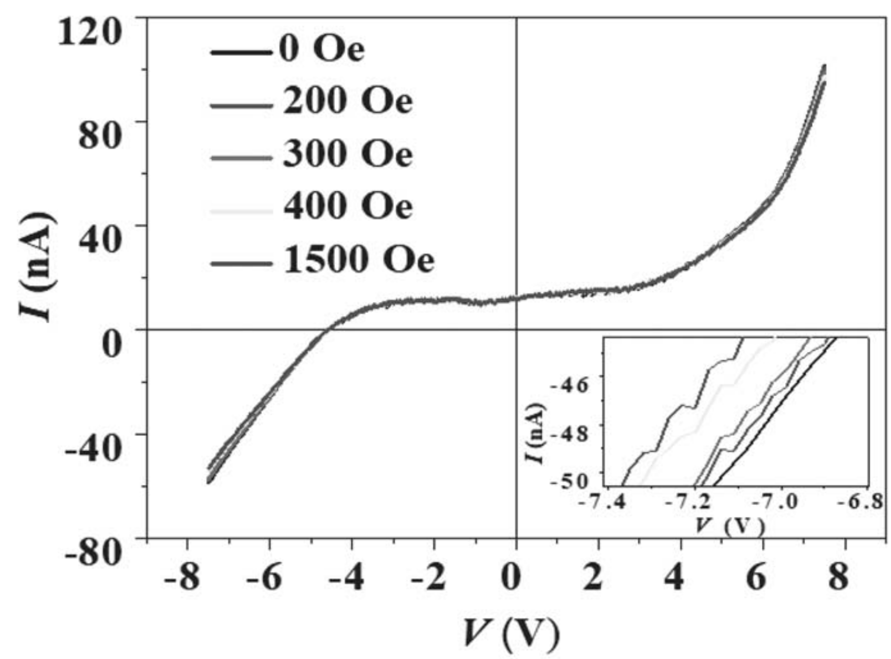

Fig.2. I-V Characteristics of p-Si/LSMO/CuPc/Au heterostructure in the presence of varying magnetic field strength. The inset shows the enlarged view of the plot in the negative bias.

and magnetic field-dependent measurements were conducted.

Results and Discussions

Fig. 1 shows the log I *vs* V plot of p-Si/$La_{0.7}$ $Sr_{0.3}MnO_3$/CuPc/Au hybrid heterostructure under -7.5 V to +7.5 V electrical bias in the absence of any illumination and magnetic field. The extent of nonlinear nature in the I-V curve may be associated with the Schottky nature of LSMO/CuPc heterojunction present within the device structure.

|ΔI| is the difference in current between two half cycles at a fixed voltage.The |ΔI| vs V plot initially exhibits an increasing trend with rising voltage. This phenomenon may stem from charge storage at various interfaces within the device. During the first half cycle, when a voltage of -7.5V is applied, the device may have undergone charging. As the voltage decreases, a point is reached where the external current direction and the internally induced current oppose each other, resulting in a complete nullification of net current. Upon further voltage decrease, at zero voltage, a current attributed to the device's charging effect is observed. Subsequent voltage increments lead to increased current, eventually reaching maximum current. A similar effect is observed in the next half cycle, with a non-zero |ΔI| indicating a memory effect within the device.

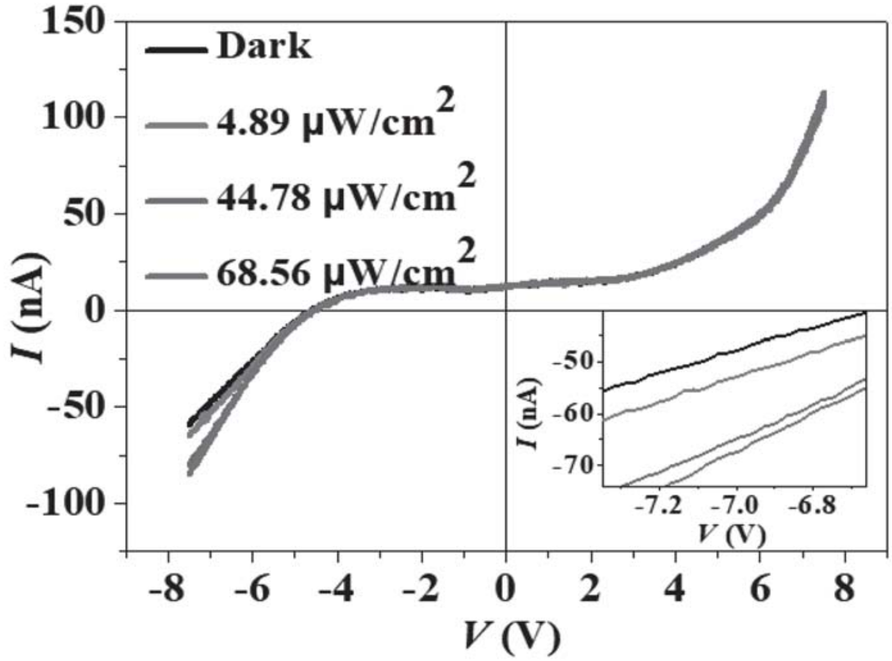

Fig.3. Light dependent I-V Characteristics of p-Si/LSMO/CuPc/Au heterostructure. Enlarged I-V characteristics in negative bias is shown in the inset.

Fig. 2 shows the I-V characteristics of the device under the variation of H. The decrement of current with increase in H is clearly observed, implying to a positive magnetoresistance (MR) response. LSMO exhibits a smaller crystal field splitting energy between the e_g and t_{2g} bands for both Mn^{3+} and Mn^{4+} valence states. The t_{2g} electrons are localized in manganites, while the localization of e_g electrons depends on the ferromagnetic double exchange between $Mn^{3+}$and Mn^{4+}valence states. In the half spin valve, the way electrons move is affected by how the energy levels of different orbitals are arranged. The dominating Jahn Teller distortion[11] in Mn3+ states lifts the degeneracy of e_g and t_{2g} orbitals. Specifically, the $e_{g2}\uparrow$ orbital has a higher energy barrier, preventing up-spin electrons from being absorbed. Consequently, down-spin electrons from LSMO fill up the $t_{2g}\downarrow$ band, experiencing larger spin-dependent scattering compared to up-spin electrons. This behaviour leads to the observed positive magnetoresistance effect.

Fig.3. illustrates the light dependent current-voltage characteristics of the half spin valve heterostructure device, with no applied magnetic field (0 Oe). The inset in the bottom right corner focuses on the curves under negative bias. Notably, it's apparent that as the power of the illuminating light increases, so does the current. It suggests that the introduction of illumination leads to the generation of a photocurrent, which amplifies as the power of the illumination increases. The 660 nm wavelength of light used falls within CuPc's absorption range[12] which may have generated excitons, mainly in the CuPc layer. Increasing light intensity enhances both exciton generation and separation at the LSMO/CuPc interface, boosting photocurrent in the device.

Conclusion

We fabricated a hybrid heterostructure p-Si/LSMO/CuPc/Au half spin valve and investigated its light and magnetic field-dependent electrical transport at room temperature. Under dark conditions, charge storage within device interfaces may have occurred, leading to a non-zero current even when 0V is applied. The non-zero |ΔI| indicates a memory effect. When varying the magnetic field, the device exhibited a positive magnetoresistance (MR) response. Additionally, changes in light intensity may have resulted in photocurrent generation within the device, which increased with higher light power.Nonlinear nature persisted indicating the Schottky nature of LSMO/CuPc heterojunction. ❐

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