Spin polarization transport in ZnO/La$_{2/3}$Sr$_{1/3}$MnO$_3$/LaAlO$_3$ heterostructures

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Abstract
ZnO/La$_{2/3}$Sr$_{1/3}$MnO$_3$ thin films have been epitaxially grown on LaAlO$_3$ (100) substrates using a pulsed laser deposition (PLD) method. I–V curves of the ZnO/La$_{2/3}$Sr$_{1/3}$MnO$_3$ structure were investigated over the temperature range from 50 to 300 K. Analysis of the leakage current demonstrates that Poole–Frenkel emission is the dominant mechanism in our sample. The photoinduced resistance and demagnetization versus temperatures demonstrates that the optical field dominated the photoconductivity mechanism below Curie temperature Tc from different green-light source. The shape and size of the barrier were changed by junction interface and interface tensile strain due to optical and magnetic external perturbations which the photoinduced characteristics modified the carrier density at the ZnO/LSMO interface. Magnetoresistance (MR) and photoinduced resistance effect is observed and the MR is related to the electron spin-dependent scattering and the interface resistance of the heterostructure. Moreover, the La$_{1-x}$Sr$_x$MnO$_3$/ZnO heterostructure exhibited a positive colossal magnetoresistance (MR) effect over the range of 50–300 K.

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Keywords
Manganese Perovskites, Photoresistance Effects, Heterojunction Films, Electronic Spins, Semiconductor ferromagnetic
I. INTRODUCTION

CMR oxides materials heterostructures that show a half-metal insulator or antiferromagnetic semiconductor ordering simultaneously are currently attracting intensive investigations due to their interesting fundamental physics as well as potential applications in information storage and spintronics. Among the REA$_{1-x}$MnO$_3$ heterostructures materials studied so far, ZnO/La$_{2/3}$Sr$_{1/3}$MnO$_3$/LaAlO heterostructures has attracted the greatest attention. If the A and RE trivalent rare earth element is partially doped in proportion with the divalent alkaline earth element, the doped manganese oxides RE$_{1-x}$A$_x$MnO$_3$ are formed and show competition coupling among electron spins, orbital order, and lattice field to exhibit photoresistance and colossal magnetoresistance (CMR) effect$^1$. The oxygen 2P orbital of colossal magnetoresistance can be explained by Zener double exchange model and Jahn-Teller effect. Heterojunction films structure of P typical manganese perovskites and N typical ZnO has the properties of ferromagnetic and semiconductor, the spin polarization determines phase inverse properties of LaSrMnO thin films and ZnO. In the past few years, due to its potential application in the quantum spins valve, robot equipment, high density storage head, sensor, and photoswitching, the research on photoresistance heterojunction thin films with the application of the external fields, such as optical field, magnetic field, temperature and electric field, including the spins dynamics and transport properties of electrons have attracted much attention$^{2-6}$. But there are very few reports about the photoresistance in heterojunction perovskite manganese oxides films. If the structure of La$_{1-x}$Sr$_x$MnO$_3$/ZnO heterojunction were fabricated with heterojunction structure and the lattice of LSMO, ZnO, and LAIO were matched, La$_2$Sr$_3$MnO$_3$/ZnO can realize quantum energy band clipping and rebuild the energy band structure at atom level with novel photoresistance and magnetoresistance properties. In this paper, based on the influence of the heterojunction of N typical ZnO and P typical La$_{1-x}$Sr$_x$MnO$_3$, Sr doping on spin-obrit coupling of doping X and structure decided Peak/Curie temperature/Neal temperature and the tolerance factor. The doped LaSrMnO$_3$ and the nanoscale La$_{0.7}$Sr$_{0.3}$MnO$_3$/ZnO multi layers heterojunction thin films are prepared on semiconductor ferromagnetic properties. The interacting mechanism between the laser pulse and heterojunction thin film of PR and MR materials is discussed in view of the interaction between the laser field and the electrons spins of small polarons. The photoresistance of heterojunction were achieved using quantum energy band and shape of potential barrier clipping to rebuilt the nanostructure film, and explore the electrons transfer and electronic spins dynamics$^{1-7}$.

II. SAMPLE PREPARATION AND PROPERTY

For the study, the La$_{2/3}$Sr$_{1/3}$MnO$_3$ and ZnO target were prepared using a sol-gel method and a solid state reaction technique. The La$_{2/3}$Sr$_{1/3}$MnO$_3$ was from the analytically pure oxides of La$_2$O$_3$, Sr(NO$_3$)$_2$, and Mn(NO$_3$)$_2$ appropriate stoichiometric
proportion of reaction method after repeated grinding and sintering at 950 °C for 24 h.
and then pressed into bulk target using 750MPa pressure which sintered at 1200 °C
for 48 h. The powder of ZnO was sintering at 500 °C for 48h from the analytically
pure oxides, and then pressed into bulk target using 650MPa pressure. The bilayer
ZnO/La$_{2/3}$Sr$_{1/3}$MnO$_3$ thin film was successively deposited on LAO substrate by a
pulsed laser deposition method (PLD) with a laser wavelength of 248 nm and a
repetition rate of 3 Hz, single pulse energy 150mJ. The LSMO thin film about 220nm
(estimated by profilometer) was deposited on LaAlO$_3$(100) using the pulse laser
deposition with the substrate 800°C, oxygen pressure 23 Pa. The film of ZnO about
70nm (estimated by profilometer) was deposited on La$_{2/3}$Sr$_{1/3}$MnO$_3$ (100), the
multilayer thin films were annealed at 800°C in oxygen pressure 6 Pa in order to get
better epitaxial character and oxygen doped deposit for 5 hours. The sample of
heterojunction thin film was placed in a JanisCCS-300 closed-circuit refrigerator
cryostat and the measured temperature range from 50k to 450k, the magnetic field of
0.1T, 0.3T, 0.5T ,0.9T were supply by the electromagnet perpendicularly applied to
the heterostructure.

Fig.1. X ray diffraction pattern of ZnO/ La$_{0.7}$Sr$_{0.3}$MnO$_3$/LAO heterostructure. The insets show the
(100) and (200) peaks of the LSMO film and LAO substrate.

The LSMO/ZnO structure is characterized by x-ray diffraction pattern shown in Fig.1.
The pattern indicates that the LSMO(100) and (200) diffraction peak occurred at
2θ =23°C and 2θ =47°C, while the ZnO (002) diffraction peak occurred at 2θ =34°C
besides the diffraction peak of LAO (100) substrate appears. The diffraction peak of
La$_{0.7}$Sr$_{0.3}$MnO$_3$/ZnO are matched with LAO substrate (lattice constant 0.38769 nm)
which the lattice constant is 0.386 and 0.379nm respectively. The results indicated
that the La$_{0.7}$Sr$_{0.3}$MnO$_3$ and ZnO thin films have better expitaxial character
respectively. The ZnO film has the structure of hexagonal awurtzite with crystal plane
orientation oriental (002) preferential epitaxial growth.

III. EXPERIMENTAL SETUP
The experimental setup and the schematic device were shown in Fig 2. The sample of
heterojunction thin film was placed in a JanisCCS-300 closed-circuit cryostat which is
temperature of 50k-300k, and magnetic field of 0.1T, 0.3T and 0.5T. The optical source for photoexcitation is Nd: YVO4 continuous wave and pulse wave laser. The power level is 40 mW, the wavelength is 532 nm, and pulse duration 300fs. The design pattern of the sample is shown in the inset of Fig. 2.

![Design Pattern](image)

Fig. 2. The design pattern of the sample ZnO/ La$_{2/3}$Sr$_{1/3}$MnO$_3$/LAO is shown

The pulse duration of modulated laser is 300fs-200 ms and the average power is 3.2 mJ/cm$^2$. The voltage signals are gathered with a Tektronix digitizing oscilloscope (100 MHz). The photoresistance and magnetoresistance were measured by the UT50 digital universal meter. The current–voltage (I–V) characteristics of the heterostructure at temperatures 220K are shown in Fig. 3. From data, we have measured I–V behavior in our ZnO/LSMO/LAO heterostructure which suggests that our heterostructure excites a lot of P-N contact dot and both electrode interfaces, bottom LSMO–ZnO and ZnO–In, are affected in a similar way. The applied field to current density curve fits with exponent relationship.

![I–V Curve](image)

Fig. 3 I–V curves of the ZnO / La$_{2/3}$Sr$_{1/3}$MnO$_3$ at 220K

The pulse voltages of different bias currents at 220 K are shown in Fig. 3. The increasing photoinduced resistivity is positive. The I-V properties of heterojunction thin film at 220K fit the exponential relationship, where the type P LSMO of heterojunction structure adopt positive electrode, type N ZnO use negative. Figure 3 shows I–V characteristics of the ZnO / La$_{2/3}$Sr$_{1/3}$MnO$_3$ substrate structure measured in
current perpendicular to plane geometry at different temperatures. We definite the current direction from the top ZnO layer to the bottom La$_{2/3}$Sr$_{1/3}$MnO$_3$ LAO layer as the positive bias, and vice versa. For the sake of clarity we have shown the I–V curves at 220 K. Apparently, the I–V curves are asymmetric, which indicates that this structure exhibits a slightly rectifying behavior. There are a large number of possible leakage current limiting mechanisms for perovskite oxides in the literature. Of all the possible mechanisms, we consider three types that are commonly observed in La$_{2/3}$Sr$_{1/3}$MnO$_3$. The first mechanism is interface-limited Schottky emission. The second is bulk-limited Poole–Frenkel emission. The third is space-charge-limited conduction. The current density for Poole–Frenkel emission is

$$J \propto \exp\left\{ -\frac{\phi - \frac{qE}{\varepsilon_0}}{k_B T} \right\}$$

where $\phi$ is the potential barrier height, $q$ is the electron charge, $E$ is the electrical field, $T$ is the absolute temperature, $\varepsilon$ is the optical frequency dielectric, and $k_B$ is the Boltzmann constant. The forward bias has small resistance. Being independent of the bias currents, all the photoinduced voltages are maximized at 130 ms. From data, we observed our LSMO/ZnO heterojunction heterostructure excites a lot of Zn loss and multilayers interfaces, LSMO–In, bottom LSMO–ZnO and ZnO–In, as well as the oxygen vacancies inject conduction electrons, which should lead to a n-type LSMO conductivity, are affected in a similar way. The high field makes the thermal electrons hop to inject to LSMO conductor bond. On the other hand, the Nordheim-Fowler type tunneling enable the carriers to inject energy order to go to P type valence band. The Schottky mechanism of conduction is generally described by

$$I = \frac{-I_{S1} I_{S2} \sinh(V_o/2kT)}{I_{S1} e x p(V_o/2kT) + I_{S2} e x p(V_o/2kT)}.$$  

IV. RESULTS AND DISCUSSION

The heterojunction resistance are defined as $\frac{dV}{dl}$ from V-I relationship and the magnetoresistance relative change are defined here as $MR = \frac{\Delta R/R = (R_H - R_O)/R_O$, where $R_H$ is the resistance of heterojunction with applied magnetic field and $R_O$ is the resistance without magnetic field. The $\Delta R/R$ is nonlinearly dependent on the temperature, and the resistance of ZnO/LSMO heterojunction at the positive bias change with temperature as depicted in Fig. 4 respectively, which the LSMO main phase transition from the metal to insulator occur at peak temperature 223K. Before the Tp, the junction resistance reduces with the decrease of temperature. Above Tp 223K, the resistance drops down with increasing temperature. Under the magnetic field 0.1T, 0.3T, and 0.5T, the magnetic resistance increase rapid respectively, as well as the curve of R-T mobiles and Tp move to lower temperature 210K direction.  


In our heterostructure, the magnetoresistance increases with magnetic field 0.1T, 0.3T and 0.5T because the active double exchange mechanics at low temperature is reduced by magnetic field and small polarons appeared. It is noted that the resistance of LSMO/ZnO is mainly from the diffuse boundary in contrast to LSMO film. Below Curie temperature Tc, the effective carriers density determined interface diffuse and is strongly effected by the component X which are decided by Sr content of half-metal manganese perovskites\textsuperscript{8,9,10,11}. With the increase of the mismatch degree at the (100) vertical orientation, the single magnetic spin of disorder reduces the extent of double exchange. As the result, the peak transition temperature Tp of metal to insular moves to the low temperature lead to resistance rate increased. Goodenough point out that the magnetic order and orbit order of cruising electron e\textsubscript{g} lead to phase competition to appear ferromagnetic to paramagnetic phase. We consider that the MR and PR phenomenon of our heterostructure is related to two factors which result from the interface diffuse of heterostructure. The first factor is derived the interface scattering and capture effect on the carrier at interface where due to the lattice mismatch at ZnO /LSMO. The second factor is that the hereojunction exist interface roughness which interface geometric fluctuation cause relaxation duration of diffuse and average period along interface orientation, and the fluctuation potential field make electrons gas scattered. Interface component in the depletion layer which leads to insulate phase and anti-parallel spins order\textsuperscript{12,13}.
Additionally, the temperature dependence of the junction resistance with applied light field and photoinduced resistance characteristics shown as Fig. 5. The temperature of LSMO layer goes up, Eg carriers hop between Mn^{3+} and Mn^{4+}, which the mobility ratio of electrons is decided by the included angle of adjacent manganese ions \( t_{\text{eff}} = \frac{1}{2} \cos \left( \frac{\theta}{2} \right) \), where the diffuse duration of coherent spin and relaxation length are affected by band length and angle on the different interface such as magnetic domain, grain boundary, and phase boundary. At low temperature, the included angle of adjacent manganese ions \( \theta \) becomes small and transfer integration increase. The spins arranged with same orientation while double exchange grows up. At high temperature, \( \theta \) becomes large and double exchange reduces. Under induced laser radiation, below Tp temperature, the optical field make the included angle of adjacent manganese ions increased. The transfer integration \( b \cos(\frac{\theta}{2}) - b \cos(\frac{\theta}{2} + \frac{\gamma}{2}) = b \sin(\frac{\delta}{2}) \sin(\frac{\theta}{2} + \frac{\gamma}{2}) \) rises up. The magnetic resistance is increased with applied optical field. Above Tp temperature, the small polarons hop actively. Double exchange reduces as well as the small polarons is formed by the lattice polar field between the upwards spins and distorted lattice, and the resistance decreased because the intense laser has little effects on spin electrons than the small polarons of hopped electric conduction. The whole curve drop down shown as Fig. 5.

V. CONCLUSIONS
In summary, ZnO/La_{0.7}Sr_{0.3}MnO_{3} heterostructure was fabricated on LAO substrate based on atom level and clipping quantum energy bond by pulsed laser ablation. This structure exhibits a slightly rectifying behavior over the temperature range 50–300 K.
Asymmetric I–V curves show the modification of the electrical characteristics of the ZnO/La$_{0.7}$Sr$_{0.3}$MnO$_3$/LAO structure. The photoresistance and magnetic resistance of ferromagnetic spin of semiconductor were dominated by charge order, orbital order, spin order, magnetic order, and lattice structure coupling from the view of energy band engineer in the ZnO/La$_{0.7}$Sr$_{0.3}$MnO$_3$ heterostructure. Additionally, we report photoresistance in ZnO/La$_{0.7}$Sr$_{0.3}$MnO$_3$ heterostructure under illumination. Our observation is attributed to the further understanding of photoresistance and positive MR and in the heterostructure and the possibility of the development and application of manganites and ZnO devices.

Acknowledgments
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