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Photoelectric effects and magnetic responses in highly rectifying Nd$_{0.7}$Sr$_{0.3}$MnO$_3$/Nb-SrTiO$_3$ heterojunctions

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Heterostructures composed of Nd$_{0.7}$Sr$_{0.3}$MnO$_3$ and Nb-SrTiO$_3$ were fabricated by laser ablation and characterized under different fields (optic, electric, and magnetic). The asymmetric ratios [$\beta = I(1V)/I(-1V)$] in I-V curves were over 10$^7$, demonstrating highly rectifying properties in a wide temperature range. Under magnetic fields, remarkable modulations of I-V curves were achieved. Significant open circuit voltages were observed when illuminated by visible lights with wavelengths of 532 nm and 650 nm. A planar back-to-back diode (Nd$_{0.7}$Sr$_{0.3}$MnO$_3$/Nb-SrTiO$_3$/Nd$_{0.7}$Sr$_{0.3}$MnO$_3$) with a good on/off ratio to visible lights was also fabricated. These results were discussed in analogy with conventional semiconductor junctions.

I. INTRODUCTION

Recently, there have been intensive studies on heterostructures composed of complex oxides.\textsuperscript{1,2} It has been expected that their properties could be more striking than those found in semiconductor heterostructures. Rectifying properties, bias tunable magnetoresistance, and photoelectric effects have been reported in perovskite-oxide heterojunctions composed of manganites and titanates.\textsuperscript{3-11} Colossal magnetoresistance manganites are typical complex oxides with strongly correlated interactions. Strontium titanate (SrTiO$_3$) is a wide-band-gap semiconductor and has good lattice match with manganites. When doped with Nb or La, SrTiO$_3$ becomes n-type conducting. Simply depositing a manganite film on an Nb doped (or La doped) SrTiO$_3$ substrate can form a single-interface junction.\textsuperscript{4} In general, these properties should strongly depend on the barrier profiles at the interface. Although the detailed descriptions of barrier profiles are hindered by the strong correlations, it is enlightening to analyze them using conventional semiconductor theories. Within such theories, energy barriers are formed at the interfaces due to the difference of Fermi levels in the materials constituting the junction.\textsuperscript{12} Based on the analysis of the I-V and C-V characteristics, Sawa \textit{et al.} concluded that the energy barriers in Pr$_{0.7}$Ca$_{0.3}$MnO$_3$/Nb-SrTiO$_3$ and La$_{0.7}$Sr$_{0.3}$MnO$_3$/Nb-SrTiO$_3$ junctions are $\sim 0.7$ and $\sim 0.6$ eV, respectively.\textsuperscript{8} Susaki \textit{et al.} found that the I-V curves of La$_{0.7}$Sr$_{0.3}$MnO$_3$/Nb-SrTiO$_3$ junctions can be well described by thermally assisted tunneling with an effectively temperature-independent Schottky barrier, which is $\sim 0.86$ eV for the oxygen deficient junction and $\sim 0.66$ eV for stoichiometric one.\textsuperscript{10} An even higher barrier height of $\sim 1.64$ eV was deduced for La$_{0.7}$Ca$_{0.3}$MnO$_3$/Nb-SrTiO$_3$ junctions by using Fowler theory to fit photoelectric experimental results.\textsuperscript{11} Despite such values of barrier height, the reported open circuit voltages $V_{OC}$ are quite small.\textsuperscript{6,9,11} In this paper, we present a study on photoelectric effects and magnetic/electric responses for highly rectifying heterojunctions composed of Nd$_{0.7}$Sr$_{0.3}$MnO$_3$ (NSMO) and 0.05 wt. % Nb-doped SrTiO$_3$ (Nb-STO). Remarkable magnetic field modulations are observed and pronounced photovoltages are achieved.

II. EXPERIMENTAL PROCEDURES

Epitaxial NSMO thin films (100 nm) were deposited on Nb-STO (001) substrates by pulsed laser deposition at 720 $^\circ$C in 1 mbar O$_2$. The grown samples were annealed at annealed temperature in oxygen of 0.5 bar for half an hour and then slowly cooled down to room temperature. As revealed by x-ray spectra, these films are of single phase and have good epitaxy. Conventional photolithography technique was used to pattern the films into areas of 1 $\times$ 1 mm$^2$. Silver (Ag) pads were evaporated onto the NSMO films and Nb-STO substrates as electrodes. The schematic view of the junctions is displayed in the inset of Fig. 1(b). Current voltage characteristics were measured using two-lead-probe method and the positive voltage applied on NSMO was defined as the forward bias. For MR measurements, the magnetic fields were applied perpendicular to the film plane. For photoelectric experiments, semiconductor laser diodes with wavelengths of 532 and 650 nm were used as light sources.

III. RESULTS AND DISCUSSIONS

Figure 1(a) presents the current voltage curves of a junction measured at different temperatures. The asymmetric ratios, $\beta = I(1V)/I(-1V)$, are over 10$^7$, demonstrating highly rectifying properties. Hysteresis is observed under negative bias voltages, but is absent under positive ones.\textsuperscript{8} A magnified view for the forward direction indicates that log$J$-V curves are essentially straight under small bias voltages and the linearity is deviated under larger bias voltages.

To further understand these junctions, detailed analyses are performed using both Schottky junction and p-n junction

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models. For either kind of junction, the $J-V$ relation in the forward direction can be expressed as $J \approx J_S \exp(qV/k_BT)$ when $qV \gg k_BT$, where $J_S$ is the saturation current density, $n$ the ideality factor, and $k_B$ the Boltzmann constant. For a perfect p-n junction or a Schottky junction with purely thermal emission process, $n$ is 1. In the studied junction, $n$ at room temperature is evaluated to be 1.1, very close to 1. This may reflect the applicability of such models. One way to deduce the energy barrier is to get the slopes of $\ln(J_S/T) - 1/T$ plot for a p-n junction and $\ln(J_S/T^2) - 1/T$ for a Schottky junction, as displayed in Fig. 1(b). These plots are almost linear above 200 K and the slopes give the energy barriers of $\sim0.7$ eV. For a Schottky diode in a pure thermionic process case, $J_S = A^*T^2 \exp(-\phi_B/k_BT)$, in which $A^*$ is effective Richardson constant and $\phi_B$ the Schottky barrier height. By setting $A^*$ to be 156 A/cm$^2K^2$ for Nb-STO, $\phi_B$ can be estimated, as given in the inset of Fig. 1(a). At room temperature, $\phi_B$ is $\sim1$ eV, much higher than those deduced using the first method. With decreasing temperature, it decreases and is reduced to $\sim0.2$ eV when the temperature is lowered to 30 K. At the same time, $n$ is increased from $\sim1.1$ at 290 K to $\sim6.6$ at 30 K. These anomalous behaviors at low temperatures are probably because the low-temperature transport is direct tunneling process $[J = J_T \exp(qV/E_0)]$. Correspondingly, at intermediate temperatures, it should be thermal assisted tunneling $[J = J_T \exp(qV/E_0), E_0 = E_{00}\coth(E_{00}/k_BT)]$. For such a process, the slope of $\ln[J_S \cosh(E_{00}/k_BT)/T] \times 1/E_0$ gives $-\phi_B$. As shown in Fig. 1(b), this plot essentially follows a straight line above 150 K and the determined barrier height is $\sim1$ eV, which is the same as that at room temperature estimated using the effective Richardson constant.

Distinct magnetic field effects were observed in these junctions. Figure 2 depicts the $I-V$ curves in the forward direction recorded at 100 K under magnetic fields of 0, 1, 3, and 5 T. Clearly, magnetoresistance $MR$, defined as $MR = 100\% \times [R(V,H) - R(V,0)]/R(V,0)$, is bias dependent. With a fixed bias voltage $V$, current $I$ increases with increasing magnetic fields, indicating negative $MR$. The maximum magnitudes of $MR$ are $\sim40\%$, $\sim54\%$, and $\sim64\%$ for the magnetic fields of 1, 3, and 5 T, respectively.

As shown in Fig. 3(a), the $I-V$ curves have clear responses to visible lights. When the junction is illuminated, carriers will be excited by incident photons, producing the photocurrents. Naively, current under illumination $I_{light}$ is simply the superposition of dark current $I_{dark}$ and photocurrent $I_L$. $I_{light} = I_{dark} + I_L$. As can be seen from Fig. 3(a), the calculated and measured light $I-V$ curves are qualitatively same. The differences at higher positive bias voltages are understandable since the barrier height would be lowered...
under a forward bias. One of the important parameters in photoelectric measurements is the open circuit voltage $V_{OC}$, which occurs when the current in the external circuit equals zero. The temperature dependences of $V_{OC}$ are presented in Fig. 3(b). $V_{OC}$ is $\sim 0.2$ V at room temperature. When the temperature is lowered to 50 K, it increases to $\sim 0.7$ V. This value is comparable to the barrier heights deduced in a previous section. Similar to $V_{OC}$, short circuit current $I_{SC}$ increases as the temperature decreases. It is of interest to study the dependences of $V_{OC}$ and $I_{SC}$ on light density $P$. It is found that a linear dependence exists between the short circuit current $I_{SC}$ and light density $P$. This is reasonable since $I_{SC}$ should be proportional to $\eta P$, where $\eta$ is the conversion efficiency. Different from $I_{SC} - P$ plot, $V_{OC}$ is proportional to $\ln P$, as displayed in the inset of Fig. 3(b). Such a dependence is the same as that in conventional p-n junctions and Schottky junctions, where $V_{OC} \sim k_B T \ln(I_L/I_S)/q$.  

To explore potential applications of such heterojunctions, a planar back-to-back diode (NSMO/Nb-STO/NSMO) was fabricated, as illustrated in the inset of Fig. 4. This piece of prototype device exhibits a good on/off ratio (over 200 at 0.5V) to light illumination. The performances under positive and negative biases are essentially symmetric. With light illumination, the photocurrent first rises with increasing voltage and then shows a saturated feature. The increase of photocurrent at low bias is probably due to the expansion of barrier region.

IV. CONCLUSIONS

NSMO/Nb-STO heterojunctions fabricated by pulsed laser deposition demonstrate excellent rectification properties, bias tunable magnetoresistance, and strong photoelectric effects. In a wide temperature range, the asymmetric ratios $[\beta = I(1V)/I(-1V)]$, are over $10^4$. In a magnetic field of 5 T, the maximum $MR$ at 100 K is $\sim 64\%$. The open circuit voltage is $\sim 0.2$ V at room temperature and increases to $\sim 0.7$ V when the temperature is lowered to 50 K. A prototype planar back-to-back diode (NSMO/Nb-STO/NSMO) with a good on/off ratio (over 200) to visible lights has also been fabricated. Conventional semiconductor theories are used to discuss these results.

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