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Exchange Bias Effect in Phase Separated $\text{La}_{0.33}\text{Pr}_{0.34}\text{Ca}_{0.33}\text{MnO}_3$ Thin Films

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Exchange bias effect is observed in the phase separated $\text{La}_{0.33}\text{Pr}_{0.34}\text{Ca}_{0.33}\text{MnO}_3$ thin films. High exchange bias field of about 1 kOe is achieved at 4 K. The exchange bias effect in $\text{La}_{0.33}\text{Pr}_{0.34}\text{Ca}_{0.33}\text{MnO}_3$ thin films might originate from the intrinsic phase separation of the $\text{La}_{0.33}\text{Pr}_{0.34}\text{Ca}_{0.33}\text{MnO}_3$ or surface effect. The dependence of exchange bias effect on temperature, cooling field, and thickness is also investigated. This work would open an avenue to the application in the magnetic memory devices based on the phase separated manganites.

Key words: Exchange bias, Phase separation, Pulse laser deposition, Manganites, Training effect

I. INTRODUCTION

Exchange bias (EB) effect has attracted intensive research attention due to its potential application in magnetic memory devices [1–3]. The typical characteristics of the EB effect is a shift in the magnetic hysteresis loop away from zero field after being cooled through the Neel temperature of the antiferromagnet in the presence of an applied magnetic field [4–6]. The occurrence of EB effect is due to the exchange interaction between antiferromagnetic (AFM) material and ferromagnetic (FM) material at their interface [4]. The EB effect has been widely studied in different coupled systems, such as FM/AFM heterostructures, FM/ferrimagnetic bilayers systems, and FM/spin glass systems [7–11]. Recently, EB effect has also been observed in perovskite manganites and cobalites with intrinsic electronic phase separation [12–17]. The interaction between the AFM phase and FM phase or spin glass phase in the phase separated manganites and cobalites gives rise to the EB effect.

$(\text{La,Pr,Ca})\text{MnO}_3$ has been proven to be a typical phase separated manganites, in which the AFM charge-ordered insulating (COI) phase and FM metallic phase coexist at submicron meter scale [18, 19]. EB effect can in principle be observed in $(\text{La,Pr,Ca})\text{MnO}_3$ systems with large scale phase separation. However, current research on the EB effect in the phase separated manganites is mainly focused on the manganites nanoparticles or nanowires systems, the EB effect on the $(\text{La,Pr,Ca})\text{MnO}_3$ films are rarely studied [20–22].

In this work, we study the observation of the EB ef-

fect on $\text{La}_{0.33}\text{Pr}_{0.34}\text{Ca}_{0.33}\text{MnO}_3$ (LPCMO) thin films. Evident EB effect with the EB field of about 1 kOe was achieved at 4 K in the 8 nm LPCMO thin films. Temperature, cooling field, and thickness dependent EB effect was also revealed in phase separated LPCMO thin films. The investigation on the EB effect in $(\text{La,Pr,Ca})\text{MnO}_3$ films not only explores the possibility of $(\text{La,Pr,Ca})\text{MnO}_3$ films as a candidate for technological application, but also helps us to understand the complex correlation coupling between FM and AFM phase.

II. EXPERIMENTS

Epitaxial LPCMO thin films with different thickness were grown on $\text{LaAlO}_3(001)$ substrates by pulsed laser deposition using a KrF excimer laser at a repetition rate of 3 Hz and laser energy density of 5.5 J/cm^2 . The growth temperature and the oxygen pressure were $700 \text{ }^\circ\text{C}$ and 40 Pa, respectively. After the film growth, the films were postannealed at the same deposition condition for half an hour, and subsequently annealed at $780 \text{ }^\circ\text{C}$ in flowing O_2 for 5 h to reduce the amount of oxygen vacancies. The magnetic measurements were carried out using the superconducting quantum interference device (SQUID) magnetometer. The magnetic field was applied parallelly to the film plane.

III. RESULTS AND DISCUSSION

Magnetization hysteresis curves for the 8 nm LPCMO thin films measured at 4 K are presented in Fig.1. In strong contrast to symmetric reversal of the magnetization in the zero field cooling (ZFC) process (Fig.1(b)),

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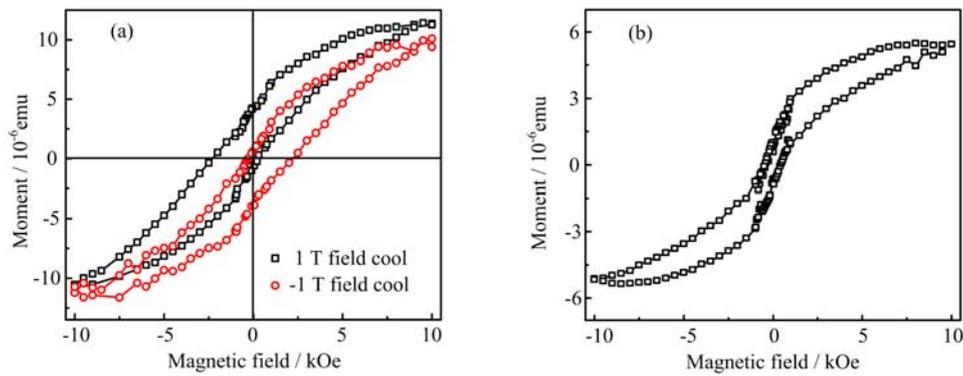


FIG. 1 (a) Magnetic hysteresis curves at 4 K for 8 nm LPCMO thin films after being field cooled from 300 K in 1 T and -1 T field. (b) Magnetic hysteresis curve at 4 K for the same LPCMO thin film after being zero field cooled from 300 K.

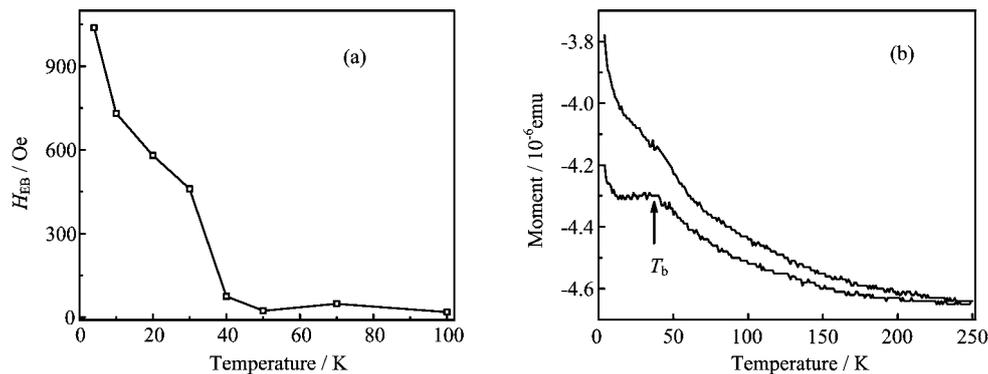


FIG. 2 (a) Temperature dependence of H_{EB} for 8 nm LPCMO thin films. (b) Temperature dependence of magnetization measured on warming with $H=200$ Oe under ZFC and FC.

the center of the magnetic hysteresis curves is shifted after being cooled in an external magnetic field of ± 10 kOe. The shift direction of the hysteresis curves is opposite to the applied cooling field. The shift of the magnetic hysteresis curves is known as EB, and the EB field was defined to be the magnitude of the center shift from zero,

$$H_{EB} = \frac{1}{2}|H_{C1} + H_{C2}| \quad (1)$$

where H_{C1} and H_{C2} are the coercive field of forward and reversed branch of the hysteresis loop, respectively [4–6]. The EB field for the 8 nm LPCMO thin films is estimated to 1.039 and 0.969 kOe for the positive- and negative-field cooling process, respectively. Such a high EB field has never been observed in the phase separated LPCMO thin films.

Figure 2 shows the EB field (H_{EB}) as a function of temperature for the 8 nm LPCMO thin films. The temperature dependent H_{EB} was measured after being field cooled in 10 kOe to the certain temperature from 250 K (Fig.2(a)). With the increase of temperature, the EB field decreases, and almost disappears above the blocking temperature $T_b \approx 40$ K (Fig.2(b)). In the phase separated manganites, the magnetic moment is frozen below

the blocking temperature, and the frozen phase is regarded as the FM metallic phase and the AFM charge ordered insulating phase in LPCMO [23, 24]. As the LPCMO thin films field cooling through T_b , a frozen interfacial exchange coupling between the FM metallic phase and AFM charge ordered insulating phase would be induced and result in the occurrence of the EB. However, when the temperature is higher than T_b , the spin of the FM cluster cannot be frozen effectively, and the EB effect disappears accordingly [21, 25].

The cooling field dependent H_{EB} was measured at 4 K, as seen in Fig.3, H_{EB} undergoes a nonlinear change, rapidly increase when the cooling field is lower than 10 kOe and decrease with the cooling field increasing further. At a low cooling field, the increase of the cooling field will align spins collinearly to the external magnetic field direction, resulting in the increase of the magnetic coupling between the interface of the FM cluster and AFM cluster, and consequently the increase of the EB field. With further increasing of the cooling field, the size of FM cluster increases as well. Analogous to the case of the FM/AFM thin films system, according to the Bean's equation, the EB field decreases while the FM layer thickness increases [4]. The decrease of the EB field at the high cooling field might be related

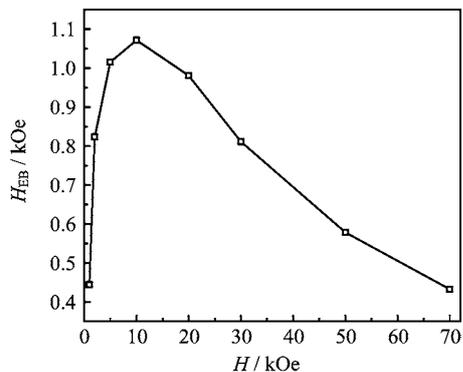


FIG. 3 H_{EB} as a function of cooling field for 8 nm LPCMO thin films.

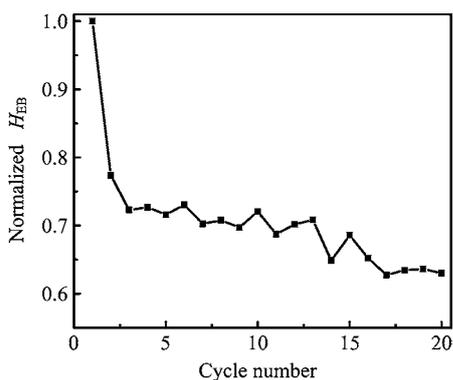


FIG. 4 Normalized H_{EB} for 8 nm LPCMO thin films as a function of the number of cycles at 4 K after being field cooled in 10 kOe.

to the increase of size of the FM cluster.

The training effect of the LPCMO thin films was also investigated at 4 K after being cooled from 250 K in 10 kOe (Fig.4). The value of H_{EB} rapidly decreases after the first cycle, and is roughly decreased with the increase of the measured cycles with some fluctuations in the subsequent cycles. The decrease of H_{EB} with increase of the cycles due to the relaxation of the interface spin configuration is in accordance with the previous studies [12, 20]. During the consecutive field cycles, the spin direction of the AFM cluster surface might be changed, which would result in a decreased exchange coupling at FM cluster and AFM cluster. Therefore, a decreased EB effect can be observed in the consecutive field cycles.

The EB effect on the phase separated LPCMO thin films may be due to the coexistence of the FM cluster and AFM cluster at the low temperature, or due to the surface effect of the thin films. It has been revealed the coexistence of the FM cluster and AFM cluster in the LPCMO thin films by adopting magnetic force microscopy [19, 26], optical conductivity [27], *etc.* The interfacial exchange coupling interaction between AFM cluster and FM cluster would result in the EB effect. It is noted that the EB effect was also observed

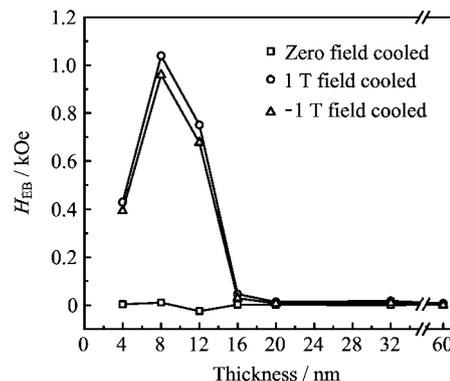


FIG. 5 Thickness dependent of H_{EB} at 4 K for LPCMO thin films after being field cooled and zero field cooled from 300 K.

in $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$ films with thickness below 3 unit cell [17], and $\text{Pr}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ film [13]. On the other hand, the surface effect becomes dominant when the size of the materials is reduced to nanoscale. However, the influence of the surface effect on the EB effect of the phase separated LPCMO thin films still needs more effort to clarify.

Thickness dependent EB effect on the LPCMO films was also investigated. As seen in Fig.5, the EB effect could only be observed for the LPCMO films with thickness below 12 nm. The thickness dependent EB effect could be interpreted in terms of phase separation between FM and AFM in LPCMO films. With the increase of thickness, the FM interaction is enhanced. The FM region increases at the expense of AFM region, resulting in a weaker AFM anisotropy [17]. When the AFM anisotropy is reduced weakly to bias FM region at the interface, EB effect can no longer be observed.

IV. CONCLUSION

EB effect is revealed in the phase separated LPCMO thin films, and the high exchange bias field of about 1 kOe is achieved at 4 K in 8 nm LPCMO thin films. The EB effect in the LPCMO thin films shows strong dependence on the temperature, cooling field and thickness. The origin of the EB effect in the LPCMO thin films is also investigated, which may be due to the intrinsic electronic phase separation or the surface effect. This work opens a new way for the application of the phase separated manganites in the technological application.

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