

Modulating Magnetism in Ferroelectric Polymer-Gated Perovskite Manganite Films with Moderate Gate Pulse Chains

Hon Fai Wong, Sheung Mei Ng, Wen Zhang, Yu Kuai Liu, Ping Kwan Johnny Wong, Chi Sin Tang, Ka Kin Lam, Xu Wen Zhao, Zhen Gong Meng, Lin Feng Fei, Wang Fai Cheng, Danny von Nordheim, Wai Yeung Wong, Zong Rong Wang, Bernd Ploss, Ji-Yan Dai, Chee Leung Mak, Andrew Thye Shen Wee, and Chi Wah Leung*

Cite This: <https://dx.doi.org/10.1021/acsami.0c14172>

Read Online

ACCESS |

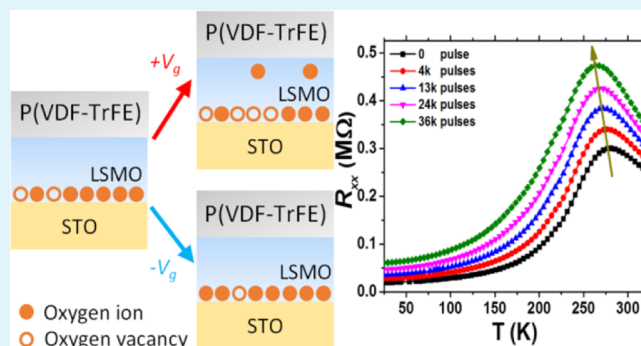
Metrics & More

Article Recommendations

Supporting Information

ABSTRACT: Most previous attempts on achieving electric-field manipulation of ferromagnetism in complex oxides, such as $\text{La}_{0.66}\text{Sr}_{0.33}\text{MnO}_3$ (LSMO), are based on electrostatically induced charge carrier changes through high- k dielectrics or ferroelectrics. Here, the use of a ferroelectric copolymer, polyvinylidene fluoride with trifluoroethylene [P(VDF-TrFE)], as a gate dielectric to successfully modulate the ferromagnetism of the LSMO thin film in a field-effect device geometry is demonstrated. Specifically, through the application of low-voltage pulse chains inadequate to switch the electric dipoles of the copolymer, enhanced tunability of the oxide magnetic response is obtained, compared to that induced by ferroelectric polarization. Such observations have been attributed to electric field-induced oxygen vacancy accumulation/depletion in the LSMO layer upon the application of pulse chains, which is supported by surface-sensitive-characterization techniques, including X-ray photoelectron spectroscopy and X-ray magnetic circular dichroism. These techniques not only unveil the electrochemical nature of the mechanism but also establish a direct correlation between the oxygen vacancies created and subsequent changes to the valence states of Mn ions in LSMO. These demonstrations based on the pulsing strategy can be a viable route equally applicable to other functional oxides for the construction of electric field-controlled magnetic devices.

KEYWORDS: electric-field effect, magnetism, LSMO, P(VDF-TrFE), oxygen vacancies



1. INTRODUCTION

Doping is frequently adopted for manipulating the transport and magnetic properties of complex oxides.^{1–4} Recent efforts include, for instances, electrostatic gating of charge carriers at oxide interfaces, across high- k dielectrics^{5,6} or ferroelectrics.^{5–7} Such electric field-induced modulation schemes permitted a change in the areal charge density below 10^{14} cm^{-2} . Electrolytes or ionic liquids (ILs) can achieve stronger modulation via the electric double-layer effect,^{8,9} capable of an interfacial capacitance of up to tens of $\mu\text{F cm}^{-2}$ (a change of $\sim 10^{15} \text{ cm}^{-2}$ in carrier density).⁹ Such a modulation has been shown to remain effective even with the gate voltage retracted,¹⁰ which arises because of the redox reactions between ILs and the channel layer.^{11–15} However, this may necessitate a protective layer in some cases.¹⁶ The strong modulation effect by ILs has facilitated the observation of various exotic phenomena, such as tunable metal–insulator transition in manganites^{13,17} and modulation of superconductivity in two-dimensional layered materials.^{18,19}

Previous attempts on electric field-induced magnetism modulation in perovskite manganites rely on epitaxially grown oxide ferroelectrics.^{7,20–23} Charge carriers at the ferroelectric/manganite interfaces were manipulated through polarization switching, thus achieving changes in the transport and magnetic behavior. This scheme, however, poses practical challenges for probing the interfacial changes at the manganite layer upon ferroelectric reversal. Polyvinylidene fluoride with trifluoroethylene [P(VDF-TrFE)], a typical ferroelectric copolymer, can be a promising alternative in this regard because of its solubility in organic solvents.²⁴ This enables direct access to the interfacial changes in the magnetic perovskite by conventional surface-sensitive techniques. Be-

Received: August 6, 2020

Accepted: November 25, 2020