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Magnetoelectric coupling at the EuO/BaTiO$_3$ interface

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Magnetization modulation by ferroelectric polarization switching is reported for the ferromagnetic-ferroelectric EuO/BaTiO$_3$ heterostructure. The value of the magnetization critical exponent $\beta$ is consistent with the expected Heisenberg-like ferromagnetism of EuO and reported Curie temperature. The critical exponent is seen to decrease with increased magnetic coupling. The results are discussed in the context of data obtained earlier for epitaxial La$_{0.67}$Sr$_{0.33}$MnO$_3$/BaTiO$_3$ heterostructures, where magnetization increases and critical exponent $\beta$ also declines with ferroelectric polarization pointing away from ferromagnetic layer. The observed similarity between two systems illustrates an importance of charge doping in magnetoelectric coupling, which can be modulated by ferroelectric polarization reversal. © 2013 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4803492]
“up” to “down” with an applied voltage in excess of 400 V, in excess of the coercive voltage of the BTO films, as noted in prior work. In all cases, the La0.67Sr0.33MnO3 was used as a conducting electrode to allow electrical control of polarization in the BTO layer. In the case of the EuO/BTO heterostructure, the effect underlying LSMO electrode provided little additional magnetization, as was established through the comparative studies with the LSMO/BTO structures.

Figure 2 shows the temperature dependence of magnetization for EuO/BTO and BTO/LSMO heterostructures. The magnetization curves clearly change for both EuO (Figure 2(a)) and LSMO (Figure 2(b)) with the change in the polarization direction of the ferroelectric BTO layer. In Figure 2(a), the magnetization plots of the EuO/BTO heterostructure indicate a ferromagnetic Curie temperature (Tc) of ~70 K even in the constant magnetic field H = 200 Oe of the measurement. This value is close to the value of Tc = 69.3 K, commonly reported for EuO. This is further confirmation that our EuO is stoichiometric.

A change in the Curie temperature is expected for both EuO and LSMO with extrinsic doping, which in turn should change with ferroelectric polarization reversal. Prior studies reported an increase in Curie temperature for EuO with electron doping (n-type doping of the EuO). As seen in Figure 2, there is no significant change in the critical magnetization temperature neither for LSMO (Ref. 23) nor for EuO with polarization reversal in BTO. The EuO has the thickness of about 100 nm so that the extrinsic doping at the EuO/BTO interface is restricted to the interface region. Since EuO is an insulator, there is limited compensating charge and only a small effect of the interface on the whole EuO layer magnetization can be expected. In other words, we expect the EuO layer Tc to be barely perturbed, as discussed in detail below. However, the magnetization curves are different for two polarization FE states for both the EuO/BTO and LSMO/BTO heterostructures. For the EuO/BTO heterostructure, there should be any contribution from the LSMO layer, it should actually act to diminish the net apparent EuO magnetization change with ferroelectric polarization. The presence of the LSMO layer within the EuO/BTO heterostructure does not completely compensate or erase the magnetization gain in the EuO when the ferroelectric polarization is away from the ferromagnetic EuO layer.

For both the EuO/BTO and LSMO/BTO heterostructures, when the BTO polarization is in the Pup state (the ferroelectric polarization is away from the ferromagnetic layer), the magnetization is larger than that for the Pdown state (the polarization is towards the ferromagnetic layer). The Pup state induces positive screening charges in LSMO/BTO interface (schematically illustrated in Figure 2(b)). On the other hand, for the same Pup state, there is a negative charge at the EuO/BTO interface, as charges are hindered from flowing through insulating EuO to the interface. The net negative charges at the EuO/BTO interface produce extrinsic doping effect on EuO. Because EuO is insulating, it is very possible that the mechanisms leading to extrinsic charge doping of the ferromagnetic layer are different for LSMO/BTO and EuO/BTO systems.

It has been known that the nearest neighbor (NN) exchange interaction J1 and the next-nearest neighbor (NNN) exchange interaction J2 are the main contributions to the magnetic coupling in the EuO system. Previous studies shown that if n-type doping in EuO was introduced, the interaction J1 will increase. From Bloch’s law:}

$$M(T) = M(0) \left[ 1 - \frac{1}{2} \left( \frac{k_BT}{2S} \right)^{3/2} \right],$$

where $M(0) = N_g \mu_B S$ is the zero-temperature magnetization. From this law, it is clear that the ferroelectric polarization at the EuO/BTO interface can increase the interaction J leading to the increasing of magnetization $M(T)$, as seen in Figure 2(a).

While valid only when T is very close to $T_C$, $M(T)$ is proportional to $(T_C - T)^\beta$ and it is safe to assume that when $(T_C - T)/T_C < 1$, there is an inverse relationship between $M(T)$ and critical exponent $\beta$. Thus, if the Pup FE state leads to doping of EuO and an increase in the magnetization, due to the increase in the interaction J1, then the critical exponent $\beta$ should decrease. On the other hand, for the Pdown FE state,
while $J_1$ and the magnetization should decrease, the critical exponent $\beta$ should increase. 

To test the premise that the interface charge changes the interaction $J_1$ of the FM layer, the temperature dependence of magnetization curves was fit to the standard 3D Heisenberg Theory $0.365$ (Refs. 33 and 35) $\beta$ value, but the trend remains clear. In the EuO/BTO system, the coupling strength, the $J$ decreases due to an increase in the magnetization, which corresponds to the polarization $P_{\text{up}}$ pointing away the FE/FM interface. The results show that extrinsic charge doping, due to an adjacent ferroelectric, affects the magnetization interaction $J$ and critical exponent $\beta$ in a manner consistent with Bloch’s $T^{3/2}$ law.

For the LSMO system, the value of critical exponent $\beta$ is a subject of much debate, as summarized in Table II. Crystal quality, different chemical compositions of the manganite, and even the temperature range selected for fitting (as also seems to be the case for the EuO/BTO system) can have noticeable effects on $\beta$. For the LSMO/BTO system, our results show consistency with prior values obtained for the LSMO system, specifically with those that fit the mean field theory predictions.

The magnetization and critical exponent $\beta$ were investigated in FM/FE heterostructures of EuO/BTO and LSMO/BTO. The reversal of the ferroelectric polarization changes the magnetization due to the charge doping effect. The critical exponent, $\beta$, decreases with the increase in the magnetization, which corresponds to the polarization $P_{\text{up}}$ pointing away the FE/FM interface. The results show that extrinsic charge doping, due to an adjacent ferroelectric, affects the magnetization interaction $J$ and critical exponent $\beta$ in a manner consistent with Bloch’s $T^{3/2}$ law.

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**Table I.** Comparison of the EuO critical exponents $\beta$ obtained by different methods.

<table>
<thead>
<tr>
<th>Method</th>
<th>$\beta$</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Film</td>
<td>$0.36^a/0.43^b$</td>
<td>This work with the fitting was obtained by selecting data range about 46 K-68 K. The corresponding $T_c$ is 69.3 K for $P_{\text{up}}/P_{\text{down}}$, respectively.</td>
</tr>
<tr>
<td>Film</td>
<td>$0.39^a/0.47^b$</td>
<td>This work with the fitting was obtained by selecting data range about 46 K-70 K. The corresponding $T_c$ is 71.0 K/70.8 K for $P_{\text{up}}/P_{\text{down}}$, respectively.</td>
</tr>
<tr>
<td>Bulk</td>
<td>0.368 (Ref. 2)</td>
<td>with $R^2$ value of 0.99248/0.99225, respectively.</td>
</tr>
<tr>
<td>Bulk</td>
<td>0.366/0.378 (Ref. 3)</td>
<td>with $R^2$ value of 0.99113/0.99007, respectively.</td>
</tr>
<tr>
<td>Bulk</td>
<td>0.370 (Ref. 4)</td>
<td>with $R^2$ value of 0.99825/0.99816, respectively.</td>
</tr>
<tr>
<td>Bulk</td>
<td>0.36 (Ref. 5)</td>
<td>with $R^2$ value of 0.99503/0.99492, respectively.</td>
</tr>
<tr>
<td>Theory (Series expansions)</td>
<td>0.38 (Ref. 6)</td>
<td>with $R^2$ value of 0.99814/0.99812, respectively.</td>
</tr>
</tbody>
</table>

$^a$This work. $^b$This work with the fitting was obtained by selecting data range about 46 K-68 K. The corresponding $T_c$ is 69.6 K/69.3 K for $P_{\text{up}}/P_{\text{down}}$ with $R^2$ value of 0.99428/0.99225, respectively.

**Table II.** Comparison of the LSMO critical exponent $\beta$ obtained by different methods.

<table>
<thead>
<tr>
<th>LSMO</th>
<th>Method</th>
<th>$\beta$</th>
<th>Notes</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{La}<em>0.67\text{Sr}</em>{0.33}\text{MnO}_3$ bulk</td>
<td>Magnetization when $P_{\text{up}}$</td>
<td>0.43$^a$</td>
<td>with $R^2$ value of 0.99825/0.99816, respectively.</td>
</tr>
<tr>
<td>$\text{La}<em>0.67\text{Sr}</em>{0.33}\text{MnO}_3$ bulk</td>
<td>Magnetization when $P_{\text{down}}$</td>
<td>0.48$^a$</td>
<td>with $R^2$ value of 0.99503/0.99492, respectively.</td>
</tr>
<tr>
<td>$\text{La}<em>0.7\text{Sr}</em>{0.3}\text{MnO}_3$ bulk</td>
<td>Magnetization</td>
<td>0.37 (Ref. 35)</td>
<td>with $R^2$ value of 0.99113/0.99007, respectively.</td>
</tr>
<tr>
<td>$\text{La}<em>0.7\text{Sr}</em>{0.3}\text{MnO}_3$ bulk</td>
<td>Neutron scattering</td>
<td>0.295 (Ref. 36)</td>
<td>with $R^2$ value of 0.99825/0.99816, respectively.</td>
</tr>
<tr>
<td>$\text{La}<em>0.7\text{Sr}</em>{0.3}\text{MnO}_3$ bulk</td>
<td>Microwave absorption</td>
<td>0.45 (Ref. 37)</td>
<td>with $R^2$ value of 0.99825/0.99816, respectively.</td>
</tr>
<tr>
<td>$\text{La}<em>0.7\text{Sr}</em>{0.3}\text{MnO}_3$ bulk</td>
<td>Magnetization</td>
<td>0.45 (Ref. 38)</td>
<td>with $R^2$ value of 0.99825/0.99816, respectively.</td>
</tr>
<tr>
<td>$\text{La}<em>0.7\text{Sr}</em>{0.3}\text{MnO}_3$ bulk</td>
<td>Ferromagnetic antiresonance</td>
<td>0.45 (Ref. 34)</td>
<td>with $R^2$ value of 0.99825/0.99816, respectively.</td>
</tr>
</tbody>
</table>

$^a$This work.
32. F. Bloch, Z. Phys. 61, 206 (1930).